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# **DIELECTRIC AND RELATED PHENOMENA**

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Faculty of Textile Engineering and Environmental Protection

Textile Institute

Bielsko-Biała, Poland

The Conference is organised under the honorary Chairmanship of

**prof.dr hab. Aleksander Łuczak**

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## LOW FREQUENCY DIELECTRIC PROPERTIES OF SOME ORGANIC COMPOUNDS

G.W. Bąk <sup>#</sup> and M. Kryszewski <sup>##</sup>

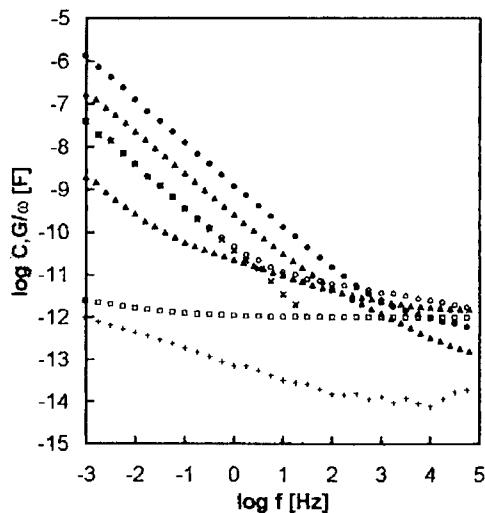
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The spectral shape of dielectric response of some solids is given by:

$$X^*(\omega) = X'(\omega) - iX''(\omega) = A_l(i\omega)^{n-1} + A_h(i\omega)^{h-1}$$

where  $X^*(\omega)$  is the complex dielectric susceptance.  $X'(\omega) = C'(\omega) - C_\infty$  and  $X''(\omega) = C''(\omega) = G(\omega)/\omega$ . The susceptance is related to the dielectric susceptibility by a geometrical factor. Low frequency dispersion (LFD) given by the equation occurs in solids in which the dielectric response is dominated by mobile charge carriers, electrons or ions. Such a type of the dielectric response has been reported to occur in chalcogenide glasses, humid ceramics and humid zeolites. Recently thorough investigations of low-frequency dielectric properties of some low-molecular weight organic materials has been carried out. LFD has been detected in some of them, i.e. in polycrystalline and amorphous tetrabenzofulvalene, polycrystalline tetracene, tianthrene, anthraquinone, fluorene and cooper phtalocyanine. It has been shown that LFD is connected with the injection of charge from electrodes. The measurements carried out on the annealed and irradiated samples of these materials and comparison with the results of thermally stimulated currents prove that the observed LFD is related to trapping and detrapping phenomena. Especially well developed LFD has been detected in iodised anthrone which is a charge transfer (CT) complex. The time-domain measurements on other CT complexes also show LFD behaviour but the LFD is not as well developed as in the case of iodised anthrone.



*Dielectric response of iodised anthrone polycrystalline pellet.*

*Fresh sample:*

$\Delta C$ ,  $\circ G/\omega$ ,  $x C-C_\infty$ .

*Annealed sample:*

$C$ ,  $+$   $G/\omega$ .

*Reiodized sample:*

$\Delta C$ ,  $\bullet G/\omega$ ,  $T=298$  K.

The results are interpreted by means of injection of charge from electrodes leading to space charge and recombination phenomena. The interpretation is a little similar to the electrochemical model of LFD proposed by Jonscher.

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## **DIELECTRIC MEASUREMENTS ON PHARMACEUTICAL MATERIALS**

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Abstract not available

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## **INSULATION LIFE**

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Abstract not available

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## OPTICAL PRE-BREAKDOWN WARNINGS IN INSULATORS

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Polymers play a significant role as insulating media because of their excellent physical and chemical properties. One of the problems encountered by these materials when they are introduced in a system is their stability versus stressing duration. The average service gradient of most insulating polymers is only a fraction ( $\approx 1/10$ ) of the breakdown strength, but space charges disturb the electrostatic field distribution. Wear-out and breakdown processes involve a dynamic situation during which the electrostatic energy stored in space charge regions is dissipated to the lattice. It is associated to the emission of visible photons, a process called electroluminescence (EL) which was observed in many different insulating polymers. Optical emissions are direct proof of excited and chemically very reactive states far from thermodynamic equilibrium. As such, they can be considered as a pre-breakdown warning and provide a probe to investigate the mechanisms leading to structural degradation.

EL has a weak intensity and its analysis necessitates the implementation of sensitive detection technics. Great care has to be taken in order to avoid electrical discharges which give rise to stray light emission. It has not been associated so far to molecular transitions of the polymer, partly because it is difficult to get wavelength resolved EL spectra, but also because of the lack of data on the nature of the luminescence centres in the investigated materials. Owing to a new experimental set-up combining integral and spectral luminescence analysis and *in situ* photoluminescence excitation, assignment of EL spectra to molecular transitions can be envisaged.

The key questions which will be addressed are : (i)-the excitation processes of the luminescent centres, (ii) their chemical nature, and (iii) the relationship between emission features and material ageing. Two types of polymers with applications in electrical engineering will be considered for the discussion:

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polyolefins which emit light through isolated chromophores situated as in chain, side-chain, or end chain groups, and aromatic polyesters with chromophores present in the repeat units that form the backbone structure of the polymer. Electroluminescence can be excited in these materials by application of d.c., a.c. and pulsed voltages. The approach relies on the analysis of charge transport process in the luminescent regime, the acquisition of electroluminescence and photoluminescence spectra, and the investigation of the field effect on photoluminescence.

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## **LIQUID CRYSTAL POLYMERS**

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Abstract not available

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## ADMITTANCE OF SURFACES WITH FRACTAL SURFACES

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The ordinary textbook expressions of electrode kinetics (including the ones of electrode admittance) have been derived by assuming some simple (e.g. planar or spherical) electrode geometry and a homogeneous current density distribution. However, in many practical cases : if e.g. rough or porous electrodes are considered, such assumptions are not justified. A possible cause of why in many cases the measured currents and impedance's deviate from the "ideal" expressions is of geometric origin : irregular e.g. rough electrode geometry can and do cause current density inhomogeneities to which the observed deviations can be attributed.

In the past decade, in connection with the peaking popularity of the concept of fractals, many theories have been constructed to find correlation between fractal electrode geometry and relevant electrochemical quantities, e.g. between the fractal dimension,  $D_f$  and the frequency dependence of the impedance. Since fractal geometry is effective in describing geometrical irregularities, the generalisation of the classical electrochemical concepts by the inclusion of fractal surfaces may provide a formalism by which the deviations might be rationalised.

In this lecture we survey the present state of this field and show the practical implications of the theories. The most important conclusions are as follows:

1. The time dependence of the diffusional flux to a fractal interface is a power law function of time, and there is a correspondence ( $\alpha = - (D_f - 1) / 2$ ) between fractal dimension and the exponent,  $\alpha$ . (Accordingly, the diffusional impedance is proportional to the power law function of frequency with the same exponent.). This equation provides a possibility for the experimental determination of the fractal dimension which provides us with the possibility that surface irregularities be characterised by a single number,  $D_f$ .
2. In principle, capacitance dispersion, due to the coupling of the solution resistance and interfacial capacitance, is expected if the electrode geometry is

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irregular. This effect can be analysed theoretically by assuming fractal geometry of the capacitive electrode: the result of these theories is that the electrode impedance is of constant phase angle (CPE) ; however, no unique relation between fractal dimension and CPE exponent can be established.

In practice, the capacitance dispersion due to the coupling of solution resistance and interfacial capacitance prevails at frequencies which are much higher than those usual in electrochemical measurements. The experimental observations of capacitance dispersion with rough electrodes can be attributed to a combined effect of the physico-chemical heterogeneity of the surface and of specific ion adsorption, as the capacitance dispersion on rough surfaces is in most cases physico-chemical rather than of purely geometrical origin.

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## **DIELECTRIC RELAXATION OF THE ELECTRIC FIELD IN POLYVINYLACETATE**

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Abstract not available

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## **DIELECTRIC RELAXATION PHENOMENA IN POLYMERS: RECENT FINDINGS BY CHEMICAL VITRIFICATION EXPERIMENTS.**

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The growing of the macromolecular chain structure during an isothermal polymerization reaction can produce similar effects to those observed in glass-forming liquids under cooling. The most prominent among these effects is the observation of the well known glass transition. However the similarities between polymerizational and thermal cooling have been not completely explored and rarely exploited to analyse the nature of co-operative interactions which produce the dielectric relaxation phenomena. Very recent experiments performed on epoxy resin systems which can be described by a bimodal dielectric relaxation, have shown that the variation of some dielectric parameters during the polymerization reaction parallels that observed in supercooling experiments. Indeed the variation of the shape parameters  $m$  and  $n$  of the low frequency relaxation change follows the predictions of the percolation theories developed for supercooled systems. Moreover, the characteristic times of the main and secondary relaxations can be described by equations which resemble the Vogel-Fulcher and Arrhenius equations, respectively. Finally, the critical exponent, by which the conductivity scales near the sol-gel transition, is in good agreement with the predictions of a percolation approach. Also, these results are confirmed for other polymeric systems, such as unsaturated polyester and polyacrylates. On the other hand, the macromolecular structure grows by a strong non linear interaction (chemical bond) among almost identical molecular units and the formed macromolecules strongly interact each other; this phenomenon has a deep similarity with that of the molecular aggregation (clusters) produced by the cooling. In conclusion the macromolecular systems seems to fulfil more accurately the theoretical predictions probably because the occurring non linear interactions with the increasing conversion are much stronger than those produced by the simply cooling of an equivalent glass-forming system.

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## **SURFACE FLOW ON INSULATORS**

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## ARROW OF TIME, PROBABILITY AND RELAXATION

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The appearance of irreversibility in physical processes and related arrow of time has raised long time ago a question about the existence of a suitable mechanism for breaking the time reversal symmetry of dynamics. It is known that not all forms of violation of time-reversal invariance can lead to the irreversibility expressed in the second law of thermodynamics. For this one needs a special form of symmetry breaking that entails a transformation from the unitary group  $(U_t)_{t \in \mathbb{R}}$  describing the reversible dynamical evolution to a Markov semigroup  $(W_t)_{t \geq 0}$  describing an irreversible evolution. The nonunitary transformation theory of irreversibility, originated over 20 years ago by Prigogine et al [1] and forwarded by others [2,3], poses the problem in the following way: does it exist a suitable operator  $\Lambda$  such that the evolution

$$\rho_O \rightarrow \Lambda U_t \rho_O = \tilde{\rho}_t$$

of the transformed state is described for  $t \geq 0$  by a strongly irreversible Markov semigroup  $W_t$ .

The possibility of time symmetry breaking is based on the existence of the so called internal time operator  $T$  introduced by Misra [2]. Operator  $T$  permits one to attribute a notion of age to individual states in such a manner that advance in internal age corresponds to increase of entropy. The explicit relationship of the internal time operator  $T$  with the time symmetry breaking transformation  $\Lambda$ , which appears to be a function of  $T$  [3], has not been clarified yet.

In order that some definite transformation could be picked out of the whole internal motion of a system, an objective criterion should exist, which allows one to distinguish between what was before the transformation and what became after it. For a given physical system, only the existence of physically different situations accessible to this system can provide such a criterion. Therefore, the notion of transformation or evolution may have a meaning only when a system can be in at least two physically different situations measured by an appropriate macroscopic

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property. It is obvious that well established relaxation data [4] provide the required criterion.

The interconnection between the probabilistic model for correlated clusters relaxation [5-7] and the Misra-Prigogine-Courbage (MPC) theory of irreversibility [3] leads to the explicit relationship between the time symmetry breaking transformation  $\Lambda$  and the internal time operator  $\mathsf{T}$

$$\Lambda = t^\alpha \mathsf{T}^{-\alpha}, \quad 0 < \alpha < 1, \quad t \leq 0$$

The crucial observation is that the mechanism for breaking the time reversal symmetry is hidden in the probabilistic meaning of the internal time operator  $\mathsf{T}$ . This result clarifies the relationship between the arrow of time and the concept of temporal fractality used for over a decade in theoretical analysis of relaxation processes in complex physical systems. It provides another basic step towards the understanding of why Lévy stable distributions are ubiquitous in nature. Moreover, it gives a promising link between the abstract MPC theory and empirical results of condensed matter physics.

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## STUDY OF $\alpha_{||}$ AND $\alpha_{\perp}$ OF CHOLESTERYL NONANOATE MOLECULE.

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The physical properties of liquid crystals have been investigated in many laboratories for a long time, because liquid crystals have found great number of applications in technics and in everyday life. These properties are connected with the shape of liquid crystal molecules. Liquid crystals are birefringent in their mesophase. Measuring refractive indices of liquid crystals one can only calculate the values of polarizability tensor components of their samples but not molecules. In this work we want to introduce the new method of calculation of polarizability tensor components of cholesteryl nonanoate molecule.

The trace of polarizability tensor  $\alpha_1 + 2\alpha_2 = \alpha_S$  has very important property because its values do not depend on the choise of the coordinate system. Therefore the trace of polarizability tensor can characterise the liquid crystal molecule. We can find more detailed information about liquid crystal molecule by the calculation of polarizability tensor components  $\alpha_{||}$  and  $\alpha_{\perp}$  of liquid crystal molecule versus the temperature. It becomes possible because we have already found the temperature dependence of order parameter of this liquid crystal. [1]. Besides in works [2-3] it was demonstrated that the order parameter  $S$  of liquid crystal molecules was proportional to the anisotropy of refractive indices ( $n_e - n_o$ ). Solving two equations you can obtain the new expressions for  $\alpha_{||}$  and  $\alpha_{\perp}$ . In this case we find:

$$\alpha_{||} = \left( \frac{M}{N} \right) \left[ \frac{n^2 - 1}{d} + 2 \left( \frac{n_e + n_o}{3k_A d} \right) \right]$$
$$\alpha_{\perp} = \left( \frac{M}{N} \right) \left[ \frac{n^2 - 1}{d} - \frac{n_e + n_o}{3k_A d} \right]$$

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where  $M$  molecular Wight,  $N$  Avogadro number,  $d$  density of liquid crystal  
 $n^2 = (n_e^2 + 2n_o^2)/3$ .

From these equations you can calculate the polarizability tensor components  $\alpha_{||}$  and  $\alpha_{\perp}$  of liquid crystal molecule having the values of refractive indices  $n_e$  and  $n_o$ , density  $d$  and coefficient  $k_A$  for the examined liquid crystal. The  $\alpha_{||}$  and  $\alpha_{\perp}$  are no linear functions of mesophase temperature. Quantity  $(\alpha_{||} - \alpha_{\perp})$  is constant in all mesophase temperatures and  $(\alpha_1 - \alpha_2)/\Delta n$  must be independent on the mesophase temperatures too.

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## **THE DIELECTRIC BEHAVIOUR OF PbO - Bi<sub>2</sub>O<sub>3</sub> ELECTRET SYSTEM AT INFRALOW FREQUENCIES**

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The relation between charging or discharging currents and a.c. dielectric properties of metal /Pb<sub>2</sub>Bi<sub>6</sub>O<sub>11</sub>/ metal structure is considered by using the computer methods for calculating the loss factor from these currents. The components of loss due to the relaxation and conduction is separately determined with this technique. The dielectric constant and the dielectric conductivity are estimated as a function of frequency in the 10<sup>-5</sup>.... 10 Hz frequency region.

The dielectric information is obtained including the distribution of relaxation times, activation energy and entropy for dipole orientation, and dipole - moment values for the structural units involved. An explanation is suggested for the influence of the simultaneous action of d.c. and a.c. voltages on dipole polarisation in Pb<sub>2</sub>Bi<sub>6</sub>O<sub>11</sub>. Comparison studies of the relaxation behaviour and the electret properties of Pb<sub>2</sub>Bi<sub>6</sub>O<sub>11</sub> is given.

The phenomenological theory of transient currents derived from the photodepolarization and dark - depolarisation experiments is reported. The empirical formula is used for this purpose. It is concluded that the model provides reasonably good fit to the experimental data.

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## **ELECTRICAL PROPERTIES OF $Pb_3O_4$ PHOTOELECTRET LAYERS**

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The study is initiated in order to investigate the photoresponce of metal / $Pb_3O_4$ / metal structures. An analysis of the growth and decay of the photocurrent, photoelectric characteristics and photovoltaic studies are presented.

The Maxwell-Wagner theory and the method of the Cole-Cole plot are applied to the analysis of a.c. impedance measurements. The dielectric properties are examined over the temperature range of 100K to 450K and illumination levels up to 2000 lux. The capacitance of the sample was found to increase as much as 40% (for 350K and 80Hz). A comparative study of the photoconductivity, photoelectric results in  $Pb_3O_4$  layers indicates both trapped and free electron mechanism photodielectric effect.

The band structure and photovoltaic are considered. The photovoltaic behaviour of the  $Pb_3O_4$  is connected with effective height of surface barriers, trap states and Dember diffusion mechanism.

The influence of phase transitions and structural bonds Pb-O in  $Pb_3O_4$  on the photoelectric behaviour is discussed. It is shown that the high resistivity with density of deep traps in  $Pb_3O_4$  layers is suitable for photoelectric applications.

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## DIELECTRIC RELAXATIONS IN ORGANIC CARRIER-DOMINATED SYSTEMS

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The dielectric response in some organic low-molecular weight compounds is defined by the injection of charge from electrodes. The dielectric response of a few organic compounds (p-terphenyl, tetracene, anthraquinone, iodised anthraquinone) shows a dielectric loss peak due to the relaxation of charge among traps. Using the equation:

$$\tau^{-1} = v_{ph} e^{\frac{E_t}{kT}} \quad (1)$$

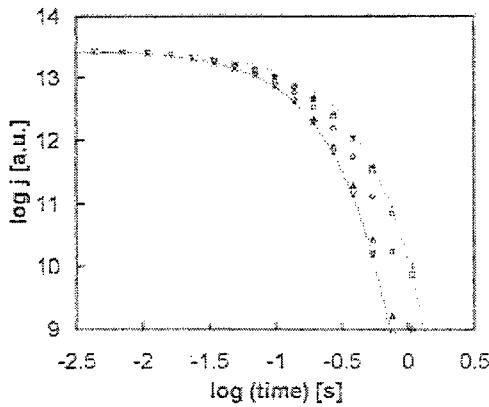
we can estimate the depth of traps provided that the concentration of traps is high enough.  $\tau$  is the relaxation time,  $v_{ph}$  is the phonon frequency,  $E_t$  is the trap depth,  $k$  is the Boltzmann constant and  $T$  is the temperature.

If the concentration of the injected charge is much greater than the concentration of the intrinsic charge than the depolarisation consists not only on the change of the spatial distribution of charge but on the outflow of the excess charge as well. If we assume that the depolarisation current is proportional to the concentration of the free charge carriers than the depolarisation process may be described by the equations:

$$\frac{dn_t}{dt} = -n_t v_{ph} e^{\frac{E_t}{kT}} + n(N_t - n_t) \sigma v \quad (2)$$

$$\frac{dn}{dt} = -\frac{dn_t}{dt} - \frac{nv}{D} \quad (3)$$

$n_t$  is the occupation of traps,  $n$  is the concentration of free carriers,  $N_t$  is the concentration of traps,  $\sigma$  is the capture cross-section of a trapping centre,  $v$  is the carrier velocity,  $D$  is the sample thickness. The numerical solutions of the equations



**Fig. 1** Depolarisation current as a function of time for various concentrations of traps.

•  $N_t = 10^{19} \text{ m}^{-3}$  x  $N_t = 10^{20} \text{ m}^{-3}$   $\Delta N_t = 10^{21} \text{ m}^{-3}$   
 o  $N_t = 10^{22} \text{ m}^{-3}$   $N_t = 10^{23} \text{ m}^{-3}$  +  $N_t = 10^{24} \text{ m}^{-3}$

The full line describes the depolarisation current given by eqn. 1 for the trap depth  $-0.65 \text{ eV}$  ( $\tau^{-1} = 13.9 \text{ s}$  resulting from eqn. 1), the dotted line results from eqn. 1 with

$$\tau^{-1} = 7.7 \text{ s.}$$

show that in the case of a single trapping level of the depth  $E_t$  the depolarisation due to the outflow of excess charge is of Debye type. The relaxation time proves to be dependent on the concentration of traps. For the concentration smaller than  $10^{21} \text{ m}^{-3}$  the relaxation time is given by eqn. 1, for the greater concentrations the relaxation time increases and becomes twice as long as for the smaller trap concentrations.

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## **PROPERTIES OF LIQUID ACETONE IN SILICA PORES: MOLECULAR DYNAMICS SIMULATION**

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Molecular dynamics simulations are reported for molecular acetone confined in a model cylindrical pore within a structure representing amorphous silica. Calculations are performed for pores of average diameters 1.5, 2.0, 2.5, and 3.0 nm. To evaluate the role of electrostatic interactions on the structure and dynamics of the confined liquid different potentials are considered: a guest molecule with and without a dipole moment; and the host material with and without electrostatic charges. A layered structure of molecules inside the pores is observed. The monolayer is always well defined but when polar forces are included the mobility of the molecules is highly restricted and localised adsorption centres can be identified. The most probable orientation of molecules on the pore walls is discussed. Translational and rotational motions within the monolayer are highly hindered and they are slower for stronger attractive potentials. Diffusion of polar molecules in the pore centre is faster than for non-polar molecules. This effect is explained in terms of increased molecular packing near the pore walls caused by strong adsorbate-adsorbent interactions.

## DENSITY OF WATER IN HIGH ELECTRIC FIELD

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Toney et al. [1] reported X-ray investigations of water at a charged Ag electrode (with a surface charge density  $\sigma_0$ ) finding that the first layer of  $H_2O$  molecules can be packed to a density about twice as large as that corresponding to the bulk water. They found it surprising as contradicting the current theories and suggested that electrostriction may be essential. We propose a *quantitative* explanation of their results by simple electrostatic and thermodynamical argument.

Let us examine if the electrostriction effect is comparable to that reported by Toney et al. (1). The electric field strength  $E$  is equal to  $\sigma_0/\epsilon_0$  ( $\epsilon_0$  - permittivity of vacuum) in the first layer of molecules at the electrode and further decays with the distance. During the process of charging, the electric field performs a work  $W$  on the first layer by reorienting the molecular dipoles.  $W$  is calculated basing on the equations of electrostatics with taking into account the dependence of the permittivity  $\epsilon = \epsilon(E, N)$  on the field and the number of molecules  $N$  as proposed recently in the problem of ions hydration [2]. The reorientation of dipoles reduces the chemical potential by  $\Delta\zeta_W$ , which is compensated by a change in the chemical potential  $\Delta\zeta_L$  due to the work [3,4]  $L$  done by the electrostriction pressure  $\Pi$  reducing the volume  $V = V(\Pi)$ . Since the actual dependence of  $V$  on  $\Pi$  is not available, we applied instead the room temperature isotherms [5]  $V = V(P)$  of water under pressure  $P$  in the absence of field. The following equation, where the l.h.s. represents  $\Delta\zeta_W$  and the r.h.s.  $\Delta\zeta_L$  respectively, has been written:

$$\frac{\partial}{\partial \varepsilon} \left( \frac{\gamma V}{\varepsilon_0} \int \sigma_0 d[\sigma_0(\varepsilon - 1)] \frac{\partial \varepsilon}{\partial N} = \int \frac{\partial V}{\partial N} dP \right)$$

and solved numerically. The derivative  $\partial \varepsilon / \partial N$  represents an expression in  $\varepsilon$  and  $\sigma_0$  calculated analytically from the implicit equation [2]:

$$\frac{3V\sigma_0(\varepsilon - n^2)}{N\mu(n^2 + 2)} = \tanh \left( \frac{\sigma_0 \varepsilon \mu(n^2 + 2)}{kT\varepsilon_0(2\varepsilon + n^2)} \right)$$

where  $n$  - refractive index,  $m$  - dipole moment,  $k$  - Boltzmann constant,  $T$  - temperature. The dependence of  $\varepsilon$  on  $\sigma_0$  has been found numerically from the latter equation and substituted to the former.

Denote  $d$  the ratio of water density in the first layer of molecules (in the field) to the density of bulk water (outside the field). From the experimental data of Toney et al. [1] obtained for two values of  $\sigma_0$  ( $\sim 25$  and  $-10 \mu\text{C Cm}^{-2}$ ) it follows  $d_{25} \approx 2.2$  and  $d_{10} \approx 1.4$ . From our calculations it follows  $d_{25} \approx 2.0$  and  $d_{10} \approx 1.5$ . The latter  $d$  values correspond to the pressures 12.5 GPa and 2.2 GPa, respectively, at which the bulk water forms a very dense ice VII phase. It does *not* mean, however, that the structure of the first layer of molecules should be identified with that of ice VII. The isotherms [5]  $V(P)$  in the integral representing  $\Delta\zeta_L$  concern liquid water under pressures from atmospheric to about 1 GPa, thereafter ice VI to about 2 GPa and further ice VII. From details of our calculation we infer that the results are almost insensitive to the volume jumps at phase transitions. Attaining such high densities becomes possible due to the aligning of the molecules along the field - we find that starting with about  $E = 1.5 \times 10^9 \text{ V/m}$  ( $\sigma_0 = 1.3 \mu\text{C cm}^{-2}$ ) all the dipole moments are parallel to the field [2].

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The evidence presented here shows that the density values of the first layer of water molecules at a charged electrode can quantitatively be explained by the effect of electrostriction.

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## **APPLICATION OF THE DISSADO-HILL THEORY OF DIELECTRIC RELAXATION TO ANALYSIS OF THE CRYSTALLISATION PROCESS IN GLASS-FORMING POLYMERS**

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The change of the  $\alpha$  relaxation process occurring during isothermal crystallisation of a glassy polymer, poly(aryl ether ketone ketone), has been recently followed [Ezquerra *et al.*, Phys. Rev. B 50, 6023 (1994)] by measurements in real time as crystallisation proceeds. We use these new experimental data and the Dissado-Hill theory to the description of the evolution of the dielectric magnitudes with crystallisation time.

## **SCALING PROPERTIES OF GLASS-FORMING LIQUIDS**

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In 1990 Dixon et al. [Phys. Rev. Lett. 65,1108 (1990)] suggested the existence of the form of universality in dielectric relaxation of glass-forming liquids. However, since then a few authors have pointed out considerable limitations and discrepancies in the proposed scaling form. We discuss several aspects of this scaling showing the origin of mentioned discrepancies and point out some new features which can lead to reconsideration of the concept of the universal scaling. Additionally, the scaling properties of several glass-forming liquids are analysed.

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## APPLICATION OF TDS FOR INVESTIGATION OF AQUEOUS PROTEIN SOLUTION

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The results of the polarization and relaxation process investigations in aqueous protein solutions by time domain dielectric spectroscopy (TDS) are presented. The dipole correlation functions (DCF) for horse myoglobin, white egg lysozyme and pancreatic ribonuclease A (RNase A) solutions in frequency range 500kHz-1 GHz was obtained in wide range of concentration, temperature and pH. Analysis of DCF of myoglobin solutions carried out in the framework of model-free approach has shown that there are several relaxation processes, described the anisotropic intermolecular motion, anisotropic Brownian tumbling and (at high concentration only) isotropic slow motion connected to electrostatic interactions of protein molecules. The latest process depends on translation diffusion of macromolecules in solution. Thus, from point of view of TDS method the solutions with concentration less than few percents are diluted and those ones just was used to investigate the protein itself at different conditions. At the investigation of pH-dependent processes in lysozyme solutions the pH of solutions ranged from 2 to 10 units. Joint data analysis, obtained from TDS and sedimentation methods was carried out. When pH of solution changing from acid to basic range the transition from monomer to dimer in the interval pH 4.5-6.5 was observed. The temperature research of RNase solution shown that by using TDS method we can observe the protein denaturation process. This process is very complex and consists of some stages. There is native state (10°-35°C), compacted denatured state, where the RNase molecules has stable in time and in temperature interval (45°-65°C} globular structure. At temperature more than 90°C the transition to stochastic coil (i.e. the process of complete denaturation) begins.

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## **DYNAMICS OF POLYMERS NEAR THE GLASS TRANSITION STUDIED BY BRILLOUIN AND DIELECTRIC SPECTROSCOPY**

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Light scattering and broadband dielectric spectroscopy have been used to investigate the relaxation processes of polyacrylates, polybutadiene and epoxy systems in a wide temperature and frequency region around the glass transition. Experimental results give evidence of a bimodal relaxation which bifurcate from a single process at temperatures higher than  $T_g$ , giving rise to a structural and secondary relaxation for temperatures approaching  $T_g$ . Polybutadiene is characterised by a bifurcation located near the glass transition, at  $T \approx 1.2 T_g$ , and the temperature behaviour of the characteristic time, of the strength, and of the shape of the relaxation function reveals that a time-temperature scaling law, as well as much of the predictions of the mode coupling theory (MCT), are fulfilled at high temperatures, where structural and secondary relaxation merge into a single process. Conversely, in the other systems the bifurcation is located at temperatures much higher than  $T_g$  remarkable deviations from MCT predictions occur and the change of the shape parameters with temperature and the general behaviour of the structural relaxation can be suitably interpreted in terms of percolative theories. These theories are appropriate also to describe the relaxation at the sol-gel transition and at the chemical vitrification of the polymerising amine/epoxy system, which were produced by the isothermal increase of connectivity. The sol-gel transition affects the d.c. conductivity, whose changes near the transition shows a power law behaviour, consistent with Flory-Stockmayer's theory.

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## TIME DOMAIN DIELECTRIC SPECTROSCOPY. THEORY AND APPLICATIONS.

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The recent appearance of Time Domain Dielectric Spectroscopy (usually called TDS) precision measuring systems may help resolve the problems of accuracy and sensitivity associated with this method when investigating different kinds of solid and liquid dielectrics. However, even in the case of two channel bridge measuring systems it is necessary to make special calibration measurements and considerable calculations in order to overcome many of the disadvantages inherent in TDS: Shortcomings in construction of coaxial lines, sampler, step generator, etc. These distortions and instabilities appear in the form of an altered shape of the incident pulse that cannot be compensated for either by the ordinary difference method or by apparatus correction of the pulse shape.

All these problems are concerned with high frequency limitations of the measuring system. On the other hand, the slow drifts of the base line and effects of truncation at a long finite time limit are primarily connected with low-frequency limitations.

Recent developments in high speed sampling techniques and digital processing, together with basic theory of TDS, have brought about a new generation of TDS measurement systems. We describe here such an instrument based on Time Domain Measuring Set (TDMS) specially designed for TDS hardware support. This system is designed for use in the measurement of dielectric parameters of liquid and solid materials over the frequency range 100 KHz to 10 GHz. The system utilises the difference method of measurement, and the registration of primary signals with nonuniform sampling. Software consists of programs of registration, accumulation

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and data collection, Fourier Analysis, Time Domain Treatment Analysis software: Fitting programs to determine spectroscopic parameters and correlation analysis in time domain.

The experimental results obtained by time domain dielectric spectroscopy for liquid (pure liquids, microemulsions, protein solutions) and solid materials (polymers, films, ferroelectrics) with different level of dc conductivity measured in different sample holders are discussed.

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## **THE SYSTEM FOR COMPLEX ELECTRIC PERMITTIVITY MEASUREMENTS OF SOLID-STATE MATERIALS IN THE RANGE OF 20 Hz TO 1 GHz**

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We worked out a fully computerised system for complex electric permittivity measurements of solid-state materials in the frequency range of 20 Hz to 1GHz and temperature from 77K to 430K. The measuring system contains three main parts:

- i) metrological
- ii) temperature controller
- iii) sample holder with specially designed Dewar and level controller of liquid nitrogen.

The metrological part is based on two Hewlett - Packard impedance analysers; HP-4284A and HP-4191A.

The temperature controller is based on modified PID system manufactured by INSTECH-Inc. controlled by PC-computer equipped with a software developed in our laboratory. The precision temperature control is of the order of 0.03K in the all range specified above. The software can realise the temperature scanning following the linear and/or step function.

Sample holder together with the level controller of liquid nitrogen and Dewar were designed and developed in our laboratory. The level of liquid nitrogen is controlled by PCcomputer with the precision of  $\pm 5\text{mm}$ .

By means of our system we can perform the measurements of complex electric permittivity as a function of temperature and frequency [ $\epsilon(i\omega, T)$ ] with much better precision then it was possible so far. In order to proof that, we will be able to present new results of  $\epsilon(i\omega, T)$  for some hexasubstituted benzene derivatives (plastic crystals). We found in these materials some evolution in relaxation mechanism which were not detected so far.

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## **LOW FREQUENCY DIELECTRIC DISPERSION AND UNIVERSAL LAW IN FERROELECTRIC POLYMERS AND IONOMERS**

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It is well known that dielectric relaxation in the majority of materials follows power laws. Mauritz [1] has recently attempted to relate the power  $n$  to (a) theories of processes that are fractal in space and time, (b) the theory of the dispersive transport of charge carriers in disordered materials, and (c) the cluster theory of the anomalous low-frequency dielectric dispersion phenomenon. It was suggested that while these related theories in themselves presently do not possess a sufficient level of microstructural detail that would permit discrimination between different materials, experimentally extracted values of  $n$  might nonetheless be used as rough qualitative indices of morphological regularity or "texture" over an array of ionic clusters or as a measure of the degree of connectivity of the overall charge pathway network.

The best disordered materials to prove this approach are polymers. In order to understand relationships between important aspects of ionic transport and the polarisation of polymers we analysed films of poly vinilidene fluoride with trifluorethylene (pure and doped by rodomine) and polymethacrylic acid R6G using dielectric spectroscopy.

I. Mauritz, K.A. *Macromolecules* 1989, **22**, 4483

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## **DYNAMIC DIELECTRIC PROPERTIES OF CARBOXYLIC ACID ESTERS IN BENZENE SOLUTION**

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Among the title substances, there is a class of molecules containing  $\text{COOR}$  ( $\text{R}=\text{alkyl}$ ) together with  $\text{OH}$  groups. These molecules offer different possibilities for hydrogen bonding and formation of clusters or aggregates, which can be studied by means of dielectric spectroscopy. Insight may be expected into the structure circumstances and dynamical processes occurring in the liquid state with those relatively complicated molecules, which to the authors' knowledge have not yet been studied dielectrically. Four molecules are regarded in comparison, containing 1 or 2  $\text{COOR}$  groups and 0 to 2  $\text{OH}$  groups, namely: (i)  $\text{S}(-)$  lactic acid methylester, (ii) succinic acid dimethylester, (iii)  $\text{L}(-)$  maleic acid dimethyl ester, (iv)  $\text{L}(+)$  tartaric acid diethylester. Their dielectric absorption was measured between 0.01 and 36 GHz at temperatures between 20 and 60°C as a function of concentration in benzene solution. The results are presented and discussed.

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## **SYNTHESIS OF TWO-DIMENSIONAL DISTRIBUTION FUNCTION OF DEFECTS IN DIELECTRIC BASED ON THERMALLY STIMULATED DEPOLARISATION DATA.**

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Electrically active defects are responsible for electret state in disordered dielectric. It might be characterised by two-dimensional distribution function according to activation energy and frequency factor.

Direct numeric reconstruction two-dimensional distribution function is to complicated for technical applications because instrumental error measuring TSD current must be no more than 0.01%.

Direct numeric reconstruction one-dimensional distribution function based on regularization algorithms requires that the instrumental errors of TSD data must not more than 3%. We suggest the numerical method reconstruction of two-dimensional distribution function based on application on family one-dimensional distribution functions for activation energy and one-dimensional distribution functions for frequency factor.

The results of suggested method applied for the reconstruction two-dimensional distribution function in PVDF films are presented.

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## **HIGH PRESSURE STUDIES OF ELECTRIC PERMITTIVITY NEAR A CRITICAL CONSULATE POINT IN BINARY SOLUTIONS**

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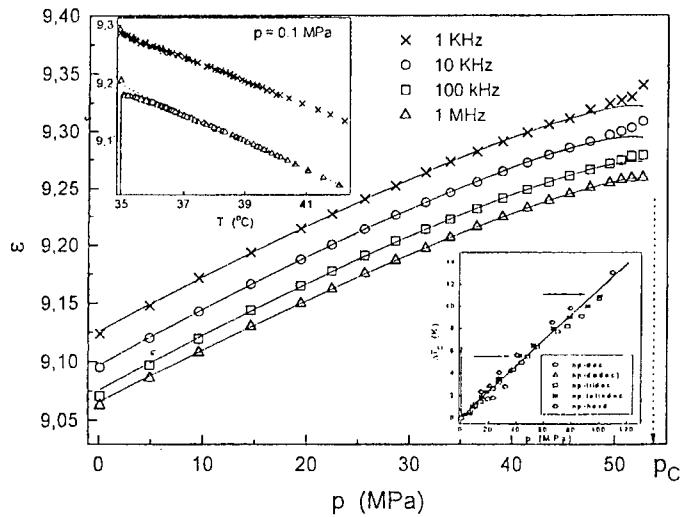
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We present, for the first time, results of studies of electric permittivity on pressure ( $p$ ) approaching the critical consolute point (CCP) in a binary solution (1-nitropropane - hexadecane and nitrobenzene - dodecane). The critical behaviour can be described by the relation isomorphic to the temperature studies under atmospheric pressure:

$$\epsilon = \epsilon_c + at + At^{1-\alpha}, \quad 1-\alpha \approx 0.89$$

with no correction to scaling terms.

In contradiction to studies under atmospheric pressure critical [1,2 and upper inset] such description is valid also for the low frequency (1 kHz). Isobaric and isothermic experiments allow to test the influence of  $dT_c/dp$  on the critical amplitude  $A$ . To test further the influence of  $dT_c/dp$  and  $dT_c/dE^2$  on the critical anomaly tests have been conducted in nitrobenzene - n - alkanes solutions with  $dT_c/dp$  varied from -0.16 K/MPa and + 0.12 K/MPa.



*Critical behaviour in 1-nitropropane hexadecane solutions. The top inset shows the behaviour under atmospheric pressure. The bottom instet tested directions of approaching the CCP.*

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## ROLE OF THE PROTON SURROUNDINGS IN FAST PROTON TRANSPORT

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It is known, that the behaviour of protons as charge carriers in solids is different than that of other ions, due to their size and absence of electron shells. Many phase transitions in crystals are related to the ordering-disordering of protons in H-bonds. Positional disorder of protons in the phase of higher symmetry can be related either to dynamical disorder between two structurally equivalent positions in H bonds (pure dielectric response on applying external electrical field) or dynamical disorder of proton subsystem between numerous structurally equivalent positions arranged in the way that the carriers can be transported through the crystal with low activation energy (diffusive-type response to external electric field).

We were interested in the role of the correlation between the fast proton migration in solids and the dynamics of the proton surrounding. Crystals of different proton surrounding  $\text{MeHSeO}_4$ ,  $\text{Me}_3\text{H}(\text{SeO}_4)_2$ ,  $\text{Me}_4\text{H}_2(\text{SeO}_4)_2$  and  $\text{Me}_4\text{LiH}_3(\text{SeO}_4)_4$  were studied. We measured:

- i) the anisotropy of electric conductivity by using the impedance spectroscopy
- ii) lattice dynamics by means of FT Raman spectroscopy
- iii) enthalpy and entropy of the superionic phase transition.

Moreover, the crystal structure in various phases was solved on the basis of X-ray diffraction. The possible mechanism of fast proton transport and the role of lattice dynamics in the family of hydrogen selenates is discussed.

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## **SIMPLE MODEL OF THE DIELECTRIC PROPERTIES OF CHOLESTERIC LIQUID CRYSTALS**

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The cholesteric liquid crystals (cholesterics) also known as chiral nematics are characterised by the arrangement in which the preferred orientation of the director  $n$  changes from layer to layer so that a helicoidal symmetry axis is formed in their volume. Only two measuring configurations are available: the "finger print", when the helix axis lies in the plane parallel to the surfaces confining the sample and "grandjean" when the helix axis is perpendicular to the surface of the sample.

It means that, with respect to the laboratory reference system, the measurements are made either in the direction parallel or perpendicular to axis of the spiral.

One could believe that the dielectric properties of the cholesterics are predictable on the basis of the physical parameters of a single molecule constituting the liquid crystal. It should imply a step forward in discussing the physical properties of this group of materials. Starting from the description in terms of "mesophase" this step would enable us to attain the level "molecule". To accomplish this project, not only the transformation principles from the liquid crystal structure frame to the molecular frame should be defined, but also a method should be offered for rendering the single molecule parameters (dipole moment  $\mu$ , and polarizability  $\alpha$ ) into a set of averaged quantities of the macroscopic ensemble ( $\epsilon \cdot n$ ). Three particular problems must be clarified before such model is effected:

1. Orientational order of the molecules of liquid crystal.
2. Definition of the local field.
3. Relation between "macroscopic parameters" ( $\epsilon \cdot n$ ) and "molecular parameters" ( $\mu, \alpha$ ).

Solution to these problems will be attempted in this study.

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## **DOUBLE POWER-LAW RELAXATION FUNCTIONS AS A WEIGHTED AVERAGE OF EXPONENTIAL RELAXATION FUNCTIONS.**

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Different types of relaxation functions, satisfying double power law, are considered. The conditions for these relaxation functions to be a weighted average of exponential relaxation functions are checked and the respective distribution of relaxation times are found to be connected with infinitely divisible laws.

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## **ELECTRIC FIELD INDUCED ROTATIONS AND RELAXATION DYNAMICS OF FLCs STUDIED BY INFRARED, DIELECTRIC AND OPTICAL METHODS**

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The IR absorbance of several groups of the LC molecules in an electrically unwound SmC\* phase was studied as a function of polariser rotation angle. The absorbance profiles under the electric field were analysed quantitatively. Field induced rotational motion of the carbonyl group and chiral C-C1 and C-CH<sub>3</sub> groups, around long molecular axis, have been confirmed to be hindered. Although the most probable orientation of C=O group is not in the plane containing the C<sub>2</sub> axis. The dynamics of the rotational motion of the molecular groups have been compared with results obtained using dielectric and optical response methods.

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## **THE DYNAMICS OF THE FERROELECTRIC LCs IN HELICAL AND SURFACE STABILISED CELLS. DIELECTRIC AND ELECTRO-OPTICAL STUDY AND A COMPUTER MODEL.**

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The dielectric and electro-optical response have been studied within the frequency range  $10^2$  -  $10^6$  Hz for a number of ferroelectric LC samples in SmC\* phase. Cells of the different thicknesses has been prepared with the bookshelf, chevron and helical structure of the FLC materials. The structure of the cells and the azimuthal angle distribution of the director have been determined by analysing optical transmission in visible region.

For surface stabilised cells, at least two peaks have been observed in dielectric and optical response spectra for temperatures far below the SmA-SmC\* transition. The first peak at the frequency of few hundreds Hz corresponds to the fluctuations of the azimuthal angle distribution across the cell (thickness mode). The relaxation time of this process depends weakly on the applied voltage, however it shows quadratic dependence on the sample thickness. The second peak, of lower frequency, is strongly dependent on the applied voltage and boundary conditions at the sample surfaces. This process most likely corresponds to the switching of the sample that reverses the direction of spontaneous polarisation vector. At low voltage the switching is initiated at defects of boundaries. With increasing of the voltage, the switching area expands as a solitary wave and involves the entire surface of the sample electrodes. Usually the process goes through a sequence of steps between different states of domains. At higher voltages (of  $0.1$  V/ $\mu$ m) the switching is becoming uniform however significant deformation of the smectic layers is observed.

A simple computer model of the ferroelectric switching, following application of sinusoidal voltage, has been proposed. Both polar and quadrupolar coupling of the liquid-crystal system to the surfaces have been considered in the model. From the numerical solution of the set of dynamic equations, an azimuthal angle modulation has been derived and then dielectric spectra and optical response has been obtained for the model.

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## **IMPROVEMENT OF DIELECTRIC PROPERTIES ON SILICON DIOXIDE SURFACES AFTER PROCESSING IN HEXAMETHYLDISILIZANE**

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Due to presence of absorbed water complexes at silicon dioxide surfaces a significant surface conductivity exists that limits application of  $\text{SiO}_2$  films as electret layers and leads to instabilities in microelectronics devices incorporating  $\text{SiO}_2$  layers. Thermal treatments and processing in hexamethyldisilizane (HMDS) are known to increase the surface resistivity of  $\text{SiO}_2$  surfaces. In this work we propose new techniques of HMDS treatment that combine the advantages of thermal and HMDS treatments.

Silicon dioxide films were obtained by oxidation of Si wafers in dry and wet oxygen. We controlled the temporal dependencies of charge at the  $\text{SiO}_2$  surfaces that is identicative of their surface conductivity. Charging was performed in the corona discharge and also with the help of a liquid scanning probe. The control of the external surface potential was done by means of a Kelvin vibrating probe. We also measured the contact angle of water droplets at the surface of  $\text{SiO}_2$  films. Measurements were carried out for the asprepared specimens and samples processed in HMDS. The distinguishing feature of our technique of HMDS processing is annealing of  $\text{SiO}_2$  films at temperatures up to 1000°C and immersion of hot specimens into HMDS. We observed correlation between the contact angle and the stability of the surface charge. The increase of hydrophobic properties resulted in better stability of this charge. In another version of HMDS treatments we stimulated the chemical reaction of HMDS decomposition at the surface of  $\text{SiO}_2$ . As a result this surface was covered with a very thin organosilicate layer and became practically hydrophobic. The obtained specimens had excellent electret properties and were thermally stable up to temperature of the order of few hundred degrees Centigrade.

## **THE USE OF LATERAL IMPEDANCE SPECTROSCOPY FOR EVALUATION OF POLYMER COATINGS IN MICROELECTRONICS**

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Encapsulation of integrated circuits is very important technical step in the microelectronics industry. The testing of polymer coatings used in microelectronics devices consists in evaluation of the effectiveness of protection of electronic circuits from moisture and vapours induced degradation. The interphase impedance is usually measured as DC leakage.

Recently impedance spectroscopy has been extensively used for transverse impedance measurements. There are only few papers on lateral impedance measurements. Of the great importance is the consideration of the influence of kind of molecules and charge carriers which determines the impedance changes.

There is a possibility to modify the microdielectrometry by plasma deposition of a selected material which acts as semipermeable membrane, serving as barrier to some liquids but absorbing moisture or particular ions. It can be shown that sensitivity less than 5 ppm of water can be obtained. The plasma deposited thin films of bromobenzene has been shown to be the appropriate coating for moisture. In the case of other gases or vapour it is possible to obtain a thin membrane which is semipermeable to the desirable species. In this way lateral impedance spectroscopy can be used to evaluate the ability of protection of encapsulation against various external conditions.

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## **HYDROPHOBIC EFFECT AND DIELECTRICAL AND OPTICAL PROPERTIES OF NONELECTROLYTE AQUEOUS SOLUTIONS**

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Our investigations of different physico-chemical properties of diluted aqueous solutions allow to suppose that hydrophobic effect is connected with quick processes in solutions. The data about dielectric properties of such systems can help to understand the nature of hydrophobic effect. But they are very poor.

In this work we analyse the concentration dependancies of some dielectric parametres for two systems: acetone ( $\text{An}$ ) -  $\text{H}_2\text{O}$  (I) and hexamethylphosphortriamid ( $\text{HMPT}$ ) -  $\text{H}_2\text{O}$  (II). These data were obtained by Chekalin ( $\text{An-H}_2\text{O}$ ) and Kesler ( $\text{HMPT-H}_2\text{O}$ ).

There are maxima in the water-rich region for concentration dependancies of relaxation amplitude of the low frequency relaxation process in (I) and nonadditive part of the dielectric permativity in (II). For interpretation of these particularities we studied the molecular light scattering in the same systems as the scattering is due to fluctuations of dielectric permativity. We found sharp additional maxima for the concentration dependancy of the intensity of light scattering in the same regions. The temperature increase decreases the amplitude of the maximum but do not change its position. Increasing of the hydrophobic part of solute molecule moves the position of the maximum in the region of more diluted solutions. As the concentration range of the maximum is very narrow one needs to have several experimental points in diluted region so as to find the position of the maximum. Such data for dielectric properties are not available now.

We consider that above mentioned maxima in the concentration dependancies of the dielectric and optical parametres for (I) and (II) systems are due to concentration fluctuations provoked by clathrate-like structure destruction and by hydrophobic interactions. They take place with the creation of the hydrates of solute molecules, out-of spheric associates and contact associates. The characteristic times of

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these processes are  $10^{-12}$ - $10^{-9}$  c. The concept of short-lived concentration fluctuations due to the above mentioned processes allows to understand the combination of dielectric and molecular light scattering data together with the data for other properties of diluted aqueous solutions.

Our investigations for aqueous solutions of acetone and HMPT show that for obtaining a full picture of hydrophobic effect it is very important to study in details static dielectric permittivity and dispersion of the dielectric permittivity of diluted nonelectrolyte solutions.

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## **DIELECTRIC RESPONSE DOMINATED BY CHARGE TRANSPORT IN THE POLY(3-NDECYLPYRROLE) FILMS.**

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We measured the d.c and the complex conductivity of poly(3-ndecylpyrrole) films in the frequency interval  $10^2$  - $10^7$  Hz in a temperatures range 300-80 K. The observed behaviour of the complex conductivity can be described by the following scaling law:

$$\frac{\sigma(\omega, T)}{\sigma(0, T)} = F \left[ \frac{A \cdot e^2 \cdot \omega}{K_B \cdot T \cdot \sigma(0, T)} \right] \quad (1)$$

Where  $F$  is the transfer function of the system, and  $\sigma(0, T)$ ,  $\sigma(\omega, T)$  are d.c and a.c. conductivity, respectively. The behaviour expressed by eq. (1) is shared by many materials, such as metal-cluster compounds, cermets, carbon-polymer composites and calcogenic glasses. In fact, all these materials have, as common property, a conductivity originated by hopping or tunnelling of the charge carriers between randomly distributed sites. Then, we can conclude that the d.c. and a.c. conductivities are originated by the same physical mechanism. Moreover, this result confirms that the charge carrier density is slightly affected by the temperature, i.e. there is no energy gap around the Fermi level.

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## THE RELAXATION OF THE NONLINEAR DIELECTRIC EFFECT

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*Nonlinear Dielectric Effect* (NDE) is based on nonlinear dependence of the electric polarisation vector on the strength of the electric field  $E$ . Experimentally NDE is observed as a very small change in electric permittivity ( $\Delta\epsilon$ ) caused by a strong, external electric field  $E$ . The NDE of polar, isotropic liquid consists of two main contributions - the negative change ( $\Delta\epsilon < 0$ ) related to orientation *Langevin effect*, and positive one ( $\Delta\epsilon > 0$ ) related to shift in molecular equilibria present in the sample (*chemical effect*). Both contributions reveal specific relaxations observed usually in separate frequency regions. Relaxation of Langevin effect takes place close to dielectric relaxation frequency ( $\omega_D$ ) and is described by specific relaxation functions [1,2].

Relaxation related with the chemical reaction is observed in two separate bands [3, 4]. The term *chemical reaction* has here a general meaning. It may describe weak dipole-dipole interactions leading to very labile molecular complexes, the intramolecular conformations and so on. The first region of chemical relaxation is associated to the rate of chemical reaction under study. The experimentally determined relaxation time  $\tau_{CH}$  can be used to calculate the rate constants of fast molecular equilibria. This part of chemical relaxation is described by pure Debye model. The second relaxation band of chemical effect occurs in dielectric relaxation region and is similar to the relaxation of Langevin effect.

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## **MORE ON COMB ELECTRODES**

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Comb electrodes are used frequently for dielectric monitoring of crosslinking reaction in epoxide adhesives. The measurements are done in frequency domain ,the electrodes are calibrated in respect to obtain reliable measurements.

However, up to knowledge of Authors there is no indication of the comparison of planar or cylindrical electrodes properties with those of comb properties performed in time domain.

Two different types of electrodes were compared with planar ones. Cu printed board etched ones with 0.2 mm spacing and ceramic laser etched with interelectrode spacing 0.04mm. The material of ceramic electrode was a mixture of noble metals. .

Plane parallel electrodes used for comparison were Cu or  $\text{SnO}_2$  coeted or printed board strips.

The comparison was done in nematic liquid crystals /NCL's/ or epoxide adhesives.

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## **EFFECT OF ELECTRIC ARC PLASMA JET TREATMENT ON ELECTRIC PROPERTIES OF MOS STRUCTURES**

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In this work the influence of electric arc plasma jet treatment (APJT) on electric parameters of Si-SiO<sub>2</sub> structures has been investigated. The Ar-air plasma flow was formed by multiple jet electric arc generator at atmospheric pressure [1].

APJT produces a hydrodynamically continuous high-enthalpy ( $>10^4$  J/g) plasma jet which impacts the surface for a short time \*(0.01-0.1 s).

The new and very interesting result of this work consists in observing the fact that carrier generation lifetime in the region outside the plasma jet treatment area and charge stability (CS) of MOS structures are abruptly diminished due to surface traps generation by UV radiation.

The test MOS-structures with dioxide width 30 nm obtained by thermal oxidation of silicon wafer in dry oxygen were used. The area of Al-electrodes was  $10^2 \div 10^3$  cm<sup>2</sup>.

To make the influence of the APJT more evident the treating plasma jet was directed along the line that lies on some distance from the centre of the wafer. It was registered the histograms of relaxation times  $\tau$  of high frequency transient capacitors and CS.

It was shown that such APJT doesn't result in significant shift of C-V ( $\Delta V_{fb} < 0.02$  V) characteristics. The most sensitive parameter of silicon subsurface region is generation time that is proportional to the  $\tau$  values. The distribution of  $\tau$  values on wafer diameter perpendicular to the treating plasma jet line is shown on Fig.1. The results show that  $\tau$  in the region outside the plasma jet treatment area is drastically diminishing due to the defect generation near Si-SiO<sub>2</sub> interface induced by near UV radiation. In the plasma jet treatment area there exists low temperature annealing of these defects for operating times less than 1 s. Two visible peaks on the figure probably arise because of doubled plasma jet used in experiments. Such  $\tau$  dependence as shown on Fig.1 indicates the existence of annealing of defects in Si

subsurface region induced by UV radiation. During the APJT UV plasma irradiation affects the structural parameters, which can be improved by athermal annealing of defects in the plasma impact area due to high density flow of plasma-excited particles ( $10^{19} \text{ cm}^{-2} \text{ s}^{-1}$ ) and hydrogen.

One of the interesting result is the correlation of  $\tau$  and CS (Fig. 2.) distributions. This is due to the influence of surface traps on both of this parameters. Thus the influence of defect on CS established with great evidence for the first time.

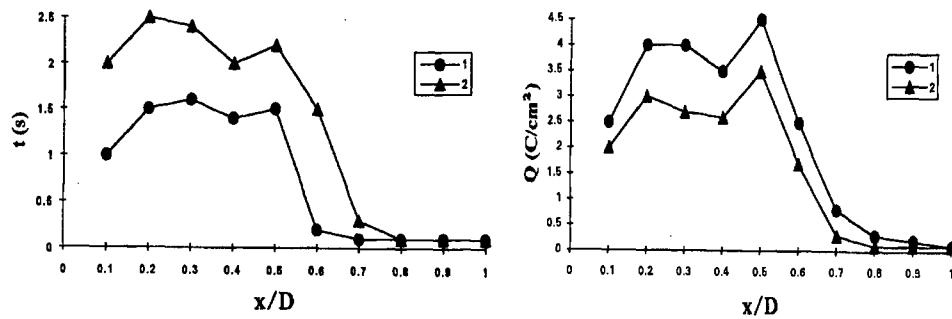


Fig.1. Distribution of  $\tau$  throughout the wafer diameter perpendicular to the treating plasma jet line for two wafers after treatment in argon plasma (dose  $10^2 \text{ W/cm}^2$  during 1 s) after hard APJT mode. Initial value of  $\tau$  was 2÷4 s (1) and 4÷6 s (2). Area of immediate contact of the plasma jet with the wafer lies in the region  $x/D=0.2\div0.5$ , where  $D=10 \text{ cm}$  is diameter of the wafer and  $x$  is the distance from the wafer border.

Fig. 2. The distribution of CS of MOS structures throughout the wafer diameter perpendicular to the treating plasma jet line for the same samples that on the Fig.1. (curves 1 and 2 corresponds curves 1 and 2 on Fig. 1, respectively) The initial  $Q$  is approximately  $0.5 \text{ C/cm}^2$

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## **SOME COMMENTS ON THE PHYSICAL SIGNIFICANCE OF THE COMPENSATION BEHAVIOUR ASSOCIATED TO THE GLASS TRANSITION RELAXATION AS OBSERVED BY THERMALLY STIMULATED CURRENTS**

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Many thermally stimulated processes obey to the so-called compensation law which is a linear relationship between the logarithm of the pre-exponential factor of the Arrhenius equation,  $\log \tau_0$ , and the apparent activation energy,  $E_a$ . Such an experimental compensation behaviour was observed in many different fields of biology, biochemistry, chemistry and physics<sup>(1,2)</sup> and has been the subject of debate during many years<sup>(3-5)</sup>. As reported by many authors, the glass transition relaxation of amorphous and semi-crystalline polymers shows a strong compensation behaviour such that there is a strong and concomitant increase of  $\tau_0$  and  $E_a$  as the glass transition temperature is approached on heating. In this context, it was suggested that the glass transition relaxation could be characterised by the so-called compensation point of coordinates  $\tau_c$ ,  $T_c$  (the compensation time and the compensation temperature), such that

$$\tau_0 = \tau_c \exp\left(-\frac{E_a}{kT_c}\right) \quad (1)$$

where  $k$  is the Bohzmann's constant. When the technique of thermally stimulated currents (t.s.c.) is used, the compensation point is thus the point of convergence of the Arrhenius lines of the thermally cleaned peaks obtained in the glass transition region. In the present communication we wish to show that the compensation point cannot really be considered as a fundamental or basic feature of the glass transition relaxation and that it is just an artefactual consequence of the strong increase of the activation energy which is observed near the glass transition temperature of

polymeric materials. In fact, if we consider the equation of a Debye peak (a thermally cleaned peak), it can be shown that it shows a maximum intensity at a temperature  $T_m$  such that

$$\frac{kT_m^2}{rE_a} = \tau_0 \exp\left(\frac{E_a}{kT_m}\right) \quad (2)$$

where  $r$  is the heating rate of the t.s.c. experiment. If we introduce now the compensation equation (1) in equation (2) we obtain

$$\frac{kT_m^2}{r\tau_c E_a} = \exp\left[\left(\frac{E_a}{k}\right)\left(\frac{1}{T_m} - \frac{1}{\tau_c}\right)\right] \quad (3)$$

which is a relationship between  $E_a$  and  $T_m$  depending only on the coordinates  $\tau_c$  and  $T_c$  of the compensation point.

The comparison of equation (3) with the experimental results obtained on different liquid crystalline polymers will show that this equation is not able to describe the glass transition relaxation in its entire temperature range, and that it is obeyed only in the particular temperature region of the glass transition relaxation where a strong increase of  $E_a$  with  $T_m$  is observed.

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## **POLARON TRANSPORT PROPERTIES DERIVED FROM MD-SIMULATED STRUCTURE IN V<sub>2</sub>O<sub>5</sub>-P<sub>2</sub>O<sub>5</sub> GLASS**

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Glasses containing transition metal ions show the „universal dynamic response” (Jonscher 1983), i.e. the ac conductivity, as a function of frequency, is given by:

$$\sigma(\omega) = \sigma(0) + A\omega^s,$$

where  $\sigma(0)$  is the dc conductivity, and the exponent  $s$  ( $\leq 1$ ) generally decreases with increasing temperature. It is found that  $s(T)$  exhibits a minimum in V<sub>2</sub>O<sub>5</sub>-P<sub>2</sub>O<sub>5</sub> glasses. This behaviour can be described in terms of overlapping polaron (OLP) tunnelling model (Long 1982). In the model the polaron hopping energy is reduced when the polaron wells at the pair of vanadium ions overlap. The power law at ac conductivity is observed in the high frequency range when the hopping occurs only between the nearest sites. Distances between vanadium atoms and their distributions have been evaluated from molecular dynamic (MD) simulations. The MD simulations, according to the Andersen algorithm (Andersen 1980), have been performed in the isobaric-isoenthalpic (NpT) ensemble. The atoms were assumed to interact by a two-body potential (Born-Mayer repulsive forces, and Coulomb forces due to full ionic charges, calculated with the aid of the Ewald technique), with Abrahamson parametrization (Abrahamson 1969). In the base of the simulation results the polaron parameters (radius, wave function decay) have been determined.

*The simulations have been performed at TASK Computer Centre (Gdansk, Poland).*

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## COMPARISON OF KWW AND POWER LAW ANALYSES IN IRON PHOSPHATE GLASSES

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Measurements of ac conductivity and discharge current in  $P_2O_5$ -FeO-MO (where M=Ca,Ba) glasses were analysed using the following procedures:

- (1) fitting the conductivity to the Jonscher power law [1], namely  $\sigma_{ac} = A\omega^s$ ,
- (2) fitting the discharge current decay to the Kohlrausch - Williams - Watt (KWW) function  $\phi(t) = \exp[-(t/r)^\beta]$ ,
- (3) estimating the stretched exponent  $\beta_m$  from the shape of the electrical modules  $M''$ . The parameter,  $\beta$ , which express the non-exponential nature of the electric current, is not substantially different if compare to the value  $\beta_s$  obtained from the frequency dependence of conductivity via the Ngai's coupling model [2] where  $\beta = 1-s$ . On the other hand a large difference is observed when compare  $\beta_s$  with  $\beta_m$  evaluated from the shape of electrical modules  $M''$ . It is shown that above discrepancy arises from the fact that  $M''$  peak is dominated by dc conductivity.

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## **DISPERSION DEPENDENCIES OF DIELECTRIC PROPERTIES. RENORMGROUP'S APPROACH.**

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The ac dielectric response of metal-insulator composites is studied numerically, using renormgroup's algorithm. For 2D and 3D random composites with bond percolation, we verify numerically that the effective dielectric function can be written numerically in the form  $\epsilon_e/\epsilon_1 = \xi^{-\tau/v} G((\epsilon_2/\epsilon_1) \xi^{(\tau+s)/v}, \xi/L)$ , where  $\epsilon_2$  and  $\epsilon_1$  are the dielectric functions,  $\xi$  is the correlation length,  $L$  is the system size (or wavelength of the electric field),  $G$  - universal functions above and below percolation,  $\tau$ ,  $s$  and  $v$  are standard percolation exponents. A similar form has been previously verified (or bond percolation by Bug *et al.* We have used a random resistor - capacitor network as a representation of a metal-insulator composite near the percolation threshold, and have calculated the scaling function using a real-space renormalization-group approach. The main idea of such approach consists of step-by-step averaging on levels of hierarchical lattices. This process is continued to fixed point of dielectric's concentration. We have found the concentration dependencies of effective characteristics of composite material for different initial concentration of dielectric in spread range of frequencies.

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## **PIEZOELECTRICITY AND DIELECTRIC PROPERTIES OF PVDF/BaTiO<sub>3</sub> COMPOSITES**

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Composite materials from polyvinylidene fluoride and ceramics barium titanate (PVDF/BaTiO<sub>3</sub>) have been prepared. Polymer/ceramic films composites with 0 - 3 connectivity pattern have been produced by introducing fine grain BaTiO<sub>3</sub> powder in the matrix of PVDF using a well-known hot-pressure method. The addition of the ceramic powder to the polymer modifies in wide range both the dielectric and the mechanical parameters which determine the piezoelectricity of the materials. These films were polarized by the conventional technique. The results of measurements of the piezoelectric parameters and the dielectric properties of the PVDF/BaTiO<sub>3</sub> mixture with the various volume fraction of the components are given in this report.

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**"IONIC TRANSPORT IN ORDER-DISORDER  
FERROELECTRICS VIA HIGH FREQUENCY DIELECTRIC  
DISPERSION AND NMR".**

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The great interest has been shown recently in power law and universality as the properties of system formed of many interacting subunits. Among such systems are solids at a "critical points" where two or more macroscopic phases become indistinguishable. It seems that quite disparate systems behave in a remarkably similar fashion near their respective critical points - because what matters most is not the details of the microscopic interactions but rather the nature of the "paths along which order is propagated" from one subunit to another distant subunit [1]. These systems also exhibit power law behaviour in their properties including dielectric respond on electromagnetic fields. So it seems that both universality and power law reflect the fact that at critical points the interaction paths between the constituent subunits in complex systems dominate the cooperative behaviour more then the detailed properties of the subunits themselves.

Based on dielectric dispersion and NMR experiments this concept will be illustrated on ferroelectrics under order - disorder phase transitions.

[1] Monthly Nature, N 12, p.41, 1995

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## **MICROSCOPIC PROPERTIES OF LOW-DIMENSIONAL MOLECULAR STRUCTURES**

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Quantum size effects, phenomena connected with low-dimensional systems, lead to some new, practically important material properties [1]. The simplest quantum structure with parameters changed only in one direction are ultrathin crystal films and superlattices [2].

The aim of this paper is to study microscopic properties of ultrathin molecular crystal films using the method of two-time, retarded, temperature dependent Green's functions [3].

We have determined the dispersion law and local densities of exciton states. Using the numerical calculations and graphical presentations the spatial distribution of elementary excitations was analysed. It is shown that excitation concentrations in the vicinity of boundary layers of films could differ from the same ones in the bulk samples.

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## **DIELECTRIC RELAXATION OF GLASS-FORMING LIQUIDS UNDER HIGH PRESSURE**

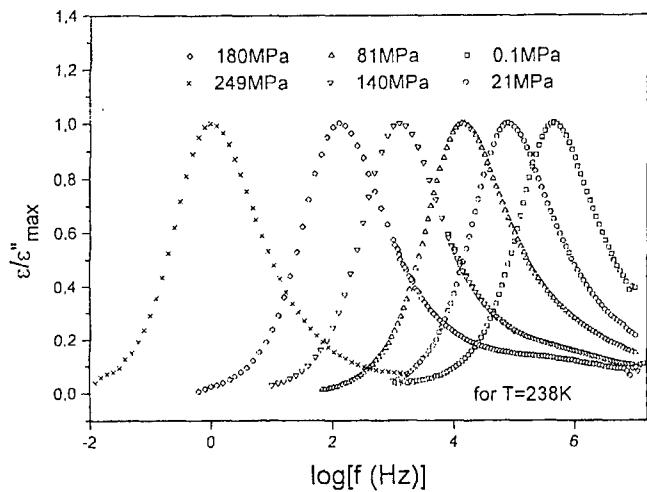
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Results of a relaxation studies in dibutyl phthalate, di-isobutyl phthalate, diethyl phthalate, dipentyl phthalate, bis(2-ethylhexyl) phthalate, di-isonyl phthalate, diallyl phthalate and dimethyl phthalate in supercooled state were performed using the dielectric spectroscopy in the frequency range from 10 mHz to 10 MHz as a function of pressure. The observed a-relaxation peaks in  $\epsilon''$  show increasing relaxation times  $\tau$  with increasing pressure at constant temperature. The figure shows normalised parts of dielectric susceptibility  $\epsilon''/\epsilon''_{\max}$  for di-isobutyl phthalate over the available frequency range for isotherm  $T=238$  K. The pressure dependence of relaxation times can be fitted by an equation

$$\tau = \tau_0 \exp(B / (p - p_0)), \quad T = \text{const}$$

where:  $B, \tau_0$  are constants, and  $p_0$  is a pressure of an ideal glass transition, which can be derived from free volume model. The correlation between shape of molecules and influence of pressure on relaxation times was noticed. Moreover the shape of absorption lines was analysed using scaling formula proposed by Dixon [1,2].



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## **MD SIMULATION STUDY OF A LIQUID CRYSTAL MODEL: GAY-BERNE PARTICLES WITH DIPOLE MOMENTS**

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Molecular dynamics simulations are reported for molecules represented by the Gay-Berne ellipsoidal particles and point dipoles perpendicular to the major axes located at the centres of mass. The system is compressed at constant temperature and the highest density corresponds to the tightest packing of hard ellipsoids. Thermodynamic, structural and dynamic properties of the system are studied and compared with available results for non-polar Gay-Berne fluid. In both system one observes transition from isotropic to smectic  $\beta$  phase. An attractive contribution to the configurational energy originating from dipole-dipole interactions is responsible for partial orientational ordering of the polar molecules for relatively low density, however, both systems reach the smectic phase at the same point. The main changes in behaviour of the polar system with respect to the non-polar one are observed in the ordered phase. The polar molecules form well defined smectic layers and the system exhibits an additional tilt transition not observed in the non-polar system. The correlation functions of translational and angular velocities and rotational correlation functions reflect observed phase transitions and show stronger restrictions of motions for the polar molecules, however, rotation of a dipole around the major axis is almost free. In the smectic phase the correlation functions of velocity component perpendicular to the major molecular axis exhibit pronounced negative parts and their shapes are analysed using a model of uncorrelated successive velocity reversals.

## **EFFECT OF ELECTRIC ARC PLASMA JET TREATMENT ON GATE OXIDE BREAKDOWN.**

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In this study, a effect of electric arc plasma jet treatment (APJT) [1] on gate oxide breakdown was investigated. The APJT modes were distinguished by exited particles flow density („soft”-  $10^{19} \text{ cm}^{-2}\text{s}^{-1}$  and „hard” -  $10^{20} \text{ cm}^{-2}\text{s}^{-1}$ ). The total dose of Ar APJT was also determined by plasma jet impacts number N (1-50). The wafers were in rotary-reciprocating motion to ensure homogeneous surface processing.

The MOS - structures consist of a Al / poly-Si / SiO<sub>2</sub>(25nm) / n-Si (4Ω cm). The gate oxide were measured by using C-V, I-V, ramped electric-field breakdown ( $E_{bd}$ ), constant current charge-to-breakdown ( $Q_{bd}$ ) and leakage voltage ( $V_l$ ) methods, DLTS.

The distributions of the initial parameters of the gate SiO<sub>2</sub> were homogeneous across the wafer surfaces and similar for all the wafers. The parameters were as follows:  $E_{bd} = 8 \text{ MV/cm}$ ;  $Q_{bd} = 0.01 \text{ to } 0.3 \text{ C}\cdot\text{cm}^{-2}$ . After APJT, the electrical parameters of the structures remained uniform across the surface. We showed that this APJT has no essential effect either on the C-V characteristics ( $\Delta V_{fb} < 0.003 \text{ V}$ ), or on DLTS parameters ( $N_{ss}$  and  $n(x)$ ). The most APJT sensitive parameter was gate oxide integrity as determined by the technique of constant current charge to breakdown (Fig.1).

Figure 1 shows typical time variation of leakage voltage  $V_l$  (the voltage on MOS structure that corresponds to a constant current of 50 μA passing through the 0.05 mm<sup>2</sup> area oxide) after APJT „soft” and „hard” modes. The initial dependence (curve 3) indicated only a slight increase in  $V_l$ , which could reach the breakdown leakage voltage  $V_{bdL}$  ( 20 V ) or breakdown leakage field  $E_{bdL}$ . The  $E_{bdL}$  is the same as

breakdown field  $E_{bd}$  measured by ramped electricfield breakdown method ( $E_{bd} = 8$  MV/cm). Figure 2 presents changes the  $E_{bd}$  against APJT impacts number. After APJT „hard” mode the leakage field increased noticeably to 30 MV/cm at first, then decreased to 12 MV/cm. After APJT „soft” mode  $E_{bd}$  dropped at first to 4 MV/cm, then grew gradually to initial value. Figure 3 presents change of the mean value  $Q_{bd}$  after APJT modes (100 chips per wafer). The initial structures had low  $Q_{bd}$  ( $0.1 \text{ C} \cdot \text{cm}^{-2}$ ). With increasing APJT dose, the fraction of chips with low  $Q_{bd}$  decreased and the fraction of high- $Q_{bd}$  ( $10 \text{ C} \cdot \text{cm}^{-2}$ ) chips increased.

The  $E_{bd}$  was diminished due to the defect generation near Si -  $\text{SiO}_2$  interface induced by plasma jet ultraviolet (UV) radiation in the case „soft” mode. After APJT „hard” mode the gate oxide had the higher quality due to higher flow density of exited particles affected the surface. Plasma exited particles flow is annealing of this UV defects and can to create a new electron traps on Si-O-H bonds. The concentration of the OH-groups with an electron capture cross-section of  $10^{-17} \text{ cm}^2$  may be as high as  $10^{17} \text{ cm}^{-3}$ . The concentration of Si-O-H defects may increase during APJT due to the quasi-chemical reactions which involve hydrogen and are induced by the transport of electron excitations generated by the plasma jet on the surface [1]. The concentration of this defects decrease with rise dose of APJT „hard” mode that results in the decrease of  $E_{bd}$  (Fig. 2). The great increase in  $Q_{bd}$  (Fig. 3) is also attributed to the electron traps evolution during APJT.

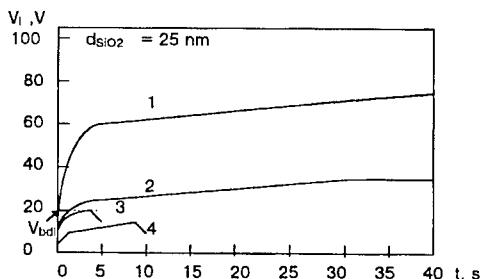


Figure 1. Temporal dependence's of  $V_d$  for MOS structure with  $d=0.1 \text{ mm}$ : (1) APJT „hard” mode,  $N=10$ , (2) APJT „hard” mode,  $N=35$ , (3) before APJT, and (4) APJT „soft” mode  $N=15$ .

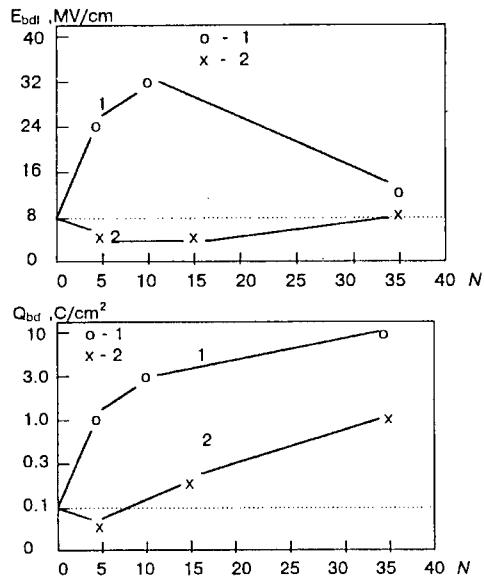


Figure 2. Leakage electric field breakdown of MOS structures for various numbers of plasma jet impacts (N). (1) APJT „hard“ mode, (2) APJT „soft“ mode.

Figure 3. Charge to breakdown of MOS structures for various numbers of plasma jet impacts (N). (1) APJT „hard“ mode, (2) APJT „soft“ mode.

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## DETERMINATION OF THE ORDER PARAMETER IN NEMATIC LIQUID CRYSTAL CONFINED TO ANOPORE MEMBRANES

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The most experimental methods using for determination of the order parameter in the nematic liquid crystal (NLC) require the sample to be macroscopically oriented. It has been shown that the confinement of NLC to inorganic filters with regular pores provides a convenient tool for the orientation of the sample in dielectric experiments [1]. The orientation of the nematic molecules inside the treated and untreated Anopore membranes allows for separate measurements of the dielectric dispersion curves for the real parts of both  $\epsilon_{\perp}$  and  $\epsilon_{\parallel}$  of the permittivity tensor. If the macroscopic molecular alignment is complete it is possible to obtain information about the order parameter from measurements of the anisotropy of electric permittivity.

The theoretical description of molecular order in the nematic phase allows us to connect the measured dielectric losses with the nematic order parameter  $S$ . The nematic order parameter calculated from the ratio of the dielectric losses connected with the slow and fast dielectric relaxation process in the 4-n-pentyl-4'-cyanobiphenyl (SCB) using Maier-Meier theory yields too low values compared with data from other experiments. However, Buka and Price [2] from measurements on a homologous compound came to a comparably low value.

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## AN INFLUENCE OF DOPING ON THE CONDUCTIVITY OF THIN DIELECTRIC LAYERS: A MONTE CARLO STUDY

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The results of extensive Monte Carlo simulation of the time-of-light (TOF) experiment for r-, and r-e-hopping transport in doped thin crystalline dielectric layers are presented. The Monte Carlo algorithm we have applied is similar to that described in Ries and Bassler (1987).

We deal with the influence of a single energetic level, which appears due to the doping (guest sites), in addition to a Gaussian distribution of hopping centres, the presence of which does not depend on doping (host sites). The dopant atoms (molecules) are considered as localised states. The jumps of the injected carriers are possible directly between the guest sites (tunnelling), and between the guest sites and the host sites (thermally activated tunnelling). In particular, we consider the energetic centres distribution of the form,  $f(e) = c_h g(\varepsilon - \varepsilon_0; \sigma) + c_g \delta(\varepsilon - \varepsilon_0 - \Delta\varepsilon)$ , where  $g(\varepsilon - \varepsilon_0; \sigma)$  is the normalised Gaussian distribution with standard deviation  $\sigma$ , centred at  $\varepsilon_0$ , and  $\delta$  is the Dirac delta function.  $c_h$  and  $c_g$  are the concentrations of host, and guest sites, respectively. The guest level is distanced by  $\Delta\varepsilon$  from  $\varepsilon_0$  (towards deeper states). The parameters common to all simulations were: the lattice constant, the Bohr radius, the external field, and the temperature.

It is shown that both r-, and r-e-hopping TOF transient currents are highly sensitive to the presence of doping level. The conductivity decreases sharply even at very low doping level ( $c_g \approx 0.01$ ), and after reaching its minimum increases again at higher doping level (e.g. Emoto and Kotani 1983). Variations of the currents shapes, and of the effective time-of-light in the function of doping are discussed. The hopping events statistics is also analysed.

*The calculations have been performed at TASK (Gdansk, Poland).*

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## **TRANSIENT CURRENTS IN THIN LAYERS FOR SIMULTANEOUS MULTIPLE-TRAPPING AND HOPPING TRANSPORT MECHANISMS**

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One of the most powerful methods for investigation the electrical carriers transport in thin layers is the classical time-of-light (TOF) experiment. The TOF measurements make it possible to determine such microscopic parameters of the investigated material, as the total trap concentration and their energetic distribution, trapping cross section, microscopic band mobility, *etc.* for multiple trapping carrier transport, and localised centres concentration, their Bohr radius, energetic distribution, *etc.*, for hopping transport. The measured TOF transient currents are usually discussed under the assumption that in the given material occurs only one of the carrier transport mechanisms, *i.e.* that the carrier transport occurs in the conductivity band, being interrupted by trapping/ detrapping events (pure multiple-trapping mechanism), or is due to pure hopping mechanism, either nearest-neighbour hopping (r-hopping), either variable-range hopping (r- $\varepsilon$ -hopping). However, multiple-trapping or hopping mechanisms are only the limit cases of the actual transport mechanism. Although under given external conditions, the carriers transport is usually dominated by one of these transport mechanisms, but it is easy to imagine the situation, where neither of the mechanisms dominates, the probability of the carrier thermal emission to the conductivity band being of the same order of magnitude as the probability of intermediate carrier tunnelling to one of the adjacent sites. Such a situation has been discussed to some extent (analytical description) by Monroe 1985 and Arkhipov and Rudenko 1987. In the present contribution we present the results of extensive Monte Carlo simulations of the TOF transient currents for the intermediate case of competing multiple-trapping and hopping transport mechanisms for spatially uniform layers with both exponential and Gaussian energetic trap distribution. Particular

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attention is paid to the temperature transition between different transport mechanisms.

*The calculations have been performed at TASK (Gdansk, Poland).*

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## THE INFLUENCE OF THE MODIFIED HOPPING CENTRE DENSITY IN THE NEAR-CONTACT REGIONS ON THE TOF CURRENTS

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We report on some new results of the Monte Carlo simulation of the time-of-light (TOF) experiment for hopping transport in non-uniformly defected crystalline thin dielectric layers. The simulation method is similar to that used in our previous works [1-3]. In the present communication the influence of the modified hopping centre distribution in the near-contact regions on the TOF transient currents is discussed. The model hopping centre distribution under consideration is:

$$N_h(x, \varepsilon) = \begin{cases} N_l \cdot f_l(\varepsilon) & 0.0 \leq x \leq D_l \\ N_b \cdot f_b(\varepsilon) & D_l < x < D_r \\ N_r \cdot f_r(\varepsilon) & D_r \leq x \leq L \end{cases}$$

where the indices l, r stay for left and right near-contact regions, and b refers to the layer bulk. As far as the energetic centres distributions  $f_i(\varepsilon)$ ,  $i=l,b,r$  are concerned, we used the normalised Gaussian distribution of standard deviation  $\sigma_i/kT$  (with  $\sigma/kT$  close to zero corresponds to a single energy level, thus the case of to r-hopping is also included. The simulations have been performed for a wide range of the parameters which appear in the above formula, covering the cases of: 1) enhanced or reduced (in respect to the layer bulk) hopping centres density in the near-contacts regions; 2) wider or narrower centres distribution in energy. The dependencies of the effective time-of flight and of the character of the current decay on the centre distribution parameters, and on the temperature are analysed.

*The simulations have been performed at TASK Computer Centre (Gdansk, Poland).*

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## **CHARGE RELAXATION AND SURFACE VOLTAGE STABILITY IN CORONA-CHARGED PTFE ELECTRETS**

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The positive excess charge relaxation in electret structures based on polytetrafluoroethylene (PTFE) films with AL deposited on one side has been investigated. Measurements were made using both the standard electret techniques and a special experimental method, based on observation of partial recovery of voltage of sandwich electret structures during reheating after forced corona-discharge. It has been found that surface voltage decay is determined largely by the process of carrier release from the surface traps, without significant recapture by deep traps in the volume of the polymer. However, in the region of the PTFE - AL interface, deep trapping occurs for carriers drifting through the bulk. Using experimental data and appropriate model, based on Simians and Tam's analysis, we determine both the energy distribution of the traps for surface and interface boundary layers. It has been shown that surface modification, particularly the treatment in polar liquids, leads to the deep traps formation in the surface density of localised states. This effect enables us to propose a series of methods for improving the surface voltage stability of PTFE - AL electret structures.

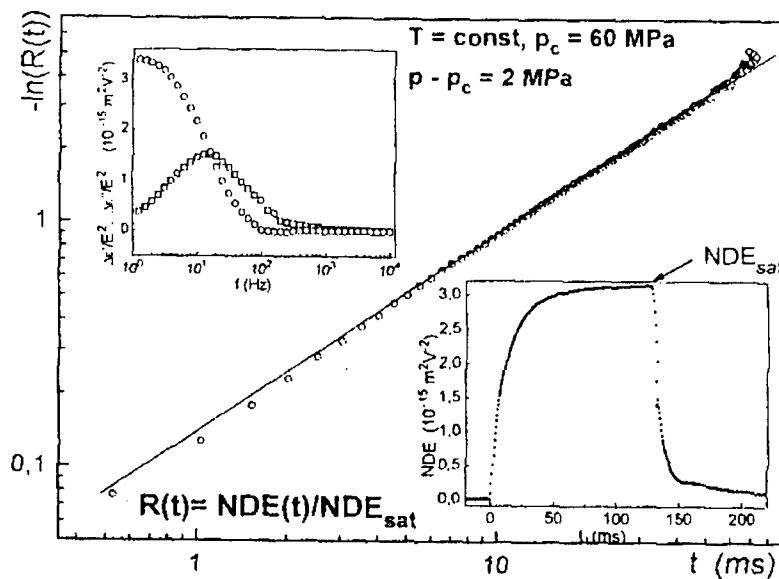
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## RELAXATION OF CRITICAL FLUCTUATIONS IN A BINARY SOLUTION. NONLINEAR DIELECTRIC EFFECT STUDIES.

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Starting from Debye's paper in 1965 [1,2,3] a vivid interest is continuously paid to a critical solution under strong electric field. In the presented research nonlinear dielectric effect (NDE) has been applied to test the stretched exponential (SE) behaviour after switching-off a strong electric field  $E$  in nitrobenzene - dodecane and 1-nitropropane - hexadecane near critical solutions. The application of high pressures allowed to reach relaxation times  $\tau \approx 0.5$  s (see Figure, 1-nitropropane - hexadecane solution) The decays has been compared with the Onuki - Doi and the Dynamic Droplet models [3]. Further new information delivered the successful transform to the frequency domain (the top inset).



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The relaxation time  $\tau$  via its relation to the measurement frequency  $f$  influences the value of the universal critical exponent  $\Psi$ :

$NDE_{sat.} = A \times (\tau - \tau_c)^{-\Psi}$  with ( $\Psi \approx 0.4$  for  $(\tau/f) > 1$ ) and ( $\Psi \approx 0.59$  for  $(\tau/f) < 1$ ). The model explaining this behaviour is proposed [4].

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## **INFLUENCE OF WATER ON THERMALLY STIMULATED CURRENT IN POROUS GLASSES**

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We have studied the influence of water absorption on electrical properties of silica porous glasses. Silica porous glasses were fabricated by leaching of two phase sodium borosilicate glasses in acid solution. The structure of porous glasses can be represented by a rigid  $\text{SiO}_2$  matrix consisting of micrometer size pores with surfaces covered with gel-like strongly hydrated  $\text{SiO}_2$ . Water can be easily absorbed and desorbed from porous glass. In order to eliminate water the samples were heated at different temperatures and processed in heksamethyldisilane (HMDS). The thermally stimulated current technique (thermally stimulated polarisation and depolarisation) was used for determination of electrical properties before and after heat and HMDS treatments. It was shown that dc conductivity and TSD peaks are attributed to the polarisation of water domains and existence of percolation net of water clusters on the walls of pores. Additionally, for the same specimens, the IR spectra recorded in the range  $500 \text{ cm}^{-1}$  to  $4000 \text{ cm}^{-1}$  were measured. The considerable differences in the IR spectra before and after heating porous glass at different temperatures were noticed.

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## **ELECTROPHYSICAL PROPERTIES OF LIQUID CRUSTAL IN DYNAMICS OF X-RAY IRRADIATION IN THE RANGE OF TEMPERATURES OF PHASE TRANSITIONS.**

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In the paper the results of X-ray irradiation on electrophysical properties in liquid crystals (LC) in the range of temperatures of phase transition smetic phase (S.L.C) - solid crystal (S.C.) are given.

It is established that doses of X-ray irradiation used ( ) do not induce destruction of liquid crystals that manifests itself in conversation after irradiation of all the temperatures of phase transitions and temperature intervals of mesophase existence. All changes of physical properties are rigorously reversible after X-ray irradiation is switched off.

It is shown that additional conductivity (where, are conductivity's of samples induced by X-ray irradiation and free from it) arising in the range if temperatures pf phase transition SLC-SC, similar to intrinsic conductivity, has peak value and is negative in sigh ( ).

In addition peaks of intrinsic and additional conductivity's are level with each other on temperature scale and peaks decrease in absolute value after multiple cleaning of LC by the method of column chromatography.

Decrease of conductivity and appearance of negative in the range of temperatures of phase transition SLC-SC in dynamics of X-ray irradiation can be explained by phenomenon of recombination of charges generated under irradiation with carriers of natural current.

Charge carriers in the range of temperatures of phase transition SLC-SC are assumed to have the same nature as in mesophase and additional conductivity is resulted from electronic mechanism of conductivity.

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## **DIELECTRIC RELAXATION IN LIQUID CRYSTAL MIXTURES (SOLUTIONS) WITH COMPONENT MOLECULES OF DIFFERENT ASYMMETRY OF SHAPE**

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In the paper the results of study of influence of chemical structure and asymmetry of molecules of substance on dielectric relaxation time, connected with dispersion of parallel component of dielectric permittivity, are given.

Liquid crystal mixtures of component molecules with different asymmetry of shape were chosen as subjects of inquiry. In this case dielectric relaxation takes place under condition of equal viscosity and degree of order for components of mixture under investigation. Hence difference in chemical structure of molecules will be the only variable parameter of the system under investigation.

It is established that Cole-Cole diagram considerably differ from Deye's one in cases of mixtures of liquid crystal substances unlike individual within the range of low frequencies.

When constructing a plot of, as a function of  $\omega$  (where  $\omega$  is imaginary part of dielectric permittivity,  $\omega$  is frequency) it is established that relaxation of, can be characterised by three dipole mechanisms with appreciably different (by an order of magnitude greater) frequency areas of dispersion.

Relaxation times, corresponding to mechanism of rotation of polar molecules components of the mixture about short transversal axis are determined in the interval of temperatures of nematic phase existence.

It is shown in that relaxation time connected with rotation of molecule about short axis depends considerably more on the length of molecules than on degree of its asymmetry.

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## **OPTICAL PROPERTIES AND MORFOLOGY OF FULLERENE [C<sub>60</sub>] LANGMUIR-BLODGETT FILMS**

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Morphology of C<sub>60</sub> fullerene Langmuir-Blodgett films having various thickness on the polycrystalline Ag surface has been studied using the surface plasmon resonance method. The complex dielectric constant of those films has been calculated. The films are shown to have a disperse structure in trough their thickness. The dispersion character is determined as depending on the surface pressure at C<sub>60</sub> layers formation on the aqueous subphase surface.

Calculated distribution functions  $S(d_i)$  are shown for single-layered and double-layered C<sub>60</sub> LB film formed at surface pressure values  $\pi=10$  and 20 mN/m. Analysis of these dependences shows that, for single-layered film and  $\pi=10$  mN/m, a significant film thickness dispersion takes place. The LB film consists mainly of 5 to 6 nm thick segments, about 3% of the substrate area remaining uncovered. For single-layered LB film formed at  $\pi= 20$  mN/m, the thickness dispersion is greater than in the first case, the  $S(d_i)$  curve being shifted towards greater thickness.

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## **DYNAMIC HETEROGENEITY IN SUPERCOOLED LIQUIDS REVEALED BY NON-RESONANT DIELECTRIC SPECTRAL HOLE BURNING**

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Only with the recent development of new experimental techniques the nature of the nonexponential relaxation of supercooled liquids and amorphous polymers has become accessible to direct study in the laboratory. These new techniques are multidimensional NMR [1], deep bleaching of fluorescent dye tracers [2], and non-resonant dielectric spectral hole burning (SHB). We will report here on the latter technique which extends the previously devised pulsed dielectric spectroscopy [3] into the large field regime. SHB can be viewed to be based on a localised dielectric heating effect and as such operates non-linearly with respect to electric field but linearly in the electric field energy.

SHB has been achieved using a large oscillating low-frequency electric field to modify the nonexponential primary dielectric relaxation of supercooled propylene carbonate and glycerol near their respective glass transitions. Observation of SHB establishes that the response is due to a distribution of relaxation times. Refilling of the spectral holes occurs exponentially in time, with a recovery rate that corresponds to the most probable relaxation time, independent of the frequency at which the response was modified. The spectral holes refill without significantly changing their shape.

The finding of a dynamic heterogeneity in the response of glass-forming materials shows that the traditionally used concept of quasi-static distributions of relaxation times breaks down. Rather than re-interpreting these distributions as effective ones, non-exponential relaxation here should be treated more properly using dynamic exchange approaches, e.g. in the spirit of environmental fluctuation models [4].

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## **AC HOPPING CONDUCTIVITY IN DISORDERED SYSTEMS : AN EXPERIMENTAL TESTING IN AMORPHOUS POLYMER THIN FILMS**

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AC conductivity in amorphous polycarbonate of Bisphenol A and polyvinylutryral thin films was obtained by dielectric measurements, in the frequency range  $10 - 10^5$  Hz at room temperature. The observed conductivity showed the typical profile for a molecular hopping systems following the power relation  $\sigma(\omega) \propto \omega^{0.97}$  in the high frequency region. This behaviour of the AC conductivity was compatible with the equation suggested by Dyres, i.e.

$$\sigma(\omega) = \sigma_0 [i\omega t_o / \ln (1 + i\omega t_o)]$$

with  $t_o = 10^{-2}$  s.

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## **RELAXATION STRUCTURAL TRANSFORMATIONS IN $\alpha\text{-As}_2\text{S}_3$ ASSOCIATED WITH RADIATION-INDUCED CENTERS**

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Structural transformations in amorphous  $\alpha\text{-As}_2\text{S}_3$  stimulated by radiation influence (accelerated electrons, gamma-quanta, neutrons etc.) are studied using differential Fourier-spectrometry technique in 400-100  $\text{cm}^{-1}$  region.

These transformations are connected with short- and intermediate-range ordering relaxation changes near specific coordination defects represented themselves the pairs of atoms with some excess of electrical charge (negative and positive) and anomalous number of covalent bonds (diamagnetic over- and under-coordinated centers introduced previously by RA. Street [1]).

It is shown that whole relaxation process is produced by the initial microstructural one-excitation of electron and (or) hole pairs localized at soft atomic configurations [2] or excitation of electron-hole pairs (excitons) [1]. The next stage includes the intermediate process-weakening of intermolecular interactions caused the possibilities to displacements of atoms and atomic blocks. The final stage of this process investigated by experimentally may be identified as destruction-polymerization transformations (switching of covalent bonds) and coordination centers stabilization in the structural host of  $\alpha\text{-As}_2\text{S}_3$  [3,4].

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# **DIELECTRIC SPECTROSCOPY OF ELECTROLYTE SOLUTIONS: A COMPARISON OF THE LOWER FREQUENCY (IONIC) RELAXATION CONTRIBUTION IN VARIOUS POLAR SOLVENTS**

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The dielectric loss spectrum of salt solutions in polar solvents consists of a conductivity and a relaxational contribution. In favourable cases, the latter can be resolved into a higher frequency contribution due to the solvent and a lower frequency contribution obviously originating in ionic aggregations. Despite of solvents and salts differing in chemical character, some common features are observed for this „ionic” spectral component. Therefore it seems questionable whether simple model assumptions, e.g. relaxation by the tumbling motion of well defined species such as ion pairs, are sufficient to account for the experimental findings in any case. It is planned to present a summary of recent results which, in comparison, may shed some light on the dynamic processes underlying the lower frequency relaxation contribution in liquid electrolytes.

## ON SOME PROCESSES IN LIQUIDCRYSTALLINE ELECTROLYTES

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Compounds capable of ionising in liquidcrystalline solvents produce electrolytic solutions. In undoped samples of liquid crystal, low-field conductance is attributed to dissociation of intrinsic impurities; but at greater fields electrode processes begin to interfere.

The conductance in the bulk depends on the dielectric constant, viscosities of the solvent, concentration and molecular dimension of the solute.

At higher concentrations ( $c \geq 10^{-4}$  moles per liter), in the low dielectric constant solvent, „clustering” takes place ( resulting in a nonlinear increase of the equivalent conductance ). In low polar solvents, like nematic liquid crystals, is high level of ionic association.

Since liquid crystals are ionic conductors, dc conduction implies a permanent exchange of charge between the molecules of the compound (dopant) and the electrodes, i.e. existence of electrochemical reactions. Electrode processes are mainly determined by electrochemical properties of the dopants (anodic or cathodic unipolar injection ).

The electrode charging transient (for enough small  $c$ ) is the ionic charge accumulation in the double layers. The kinetics of the discharge depend on diffusion from the double layers into bulk of sample ( the charge did not leave the cell via the electrodes ). The reversal of polarity causes a reversal of the potential of the double layer and the transport of the charge to the other electrode.

Conductivity-time relation has been imputed to the adsorption of ions on the glass surfaces of sample's electrodes (conductivity decrease due to adsorption phenomena because glass presents superficial polar groups Si - OH, which are able to „retain” ions by electrostatic interations ).

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## **DIELECTRIC RESPONSE OF VISCOUS AND GLASSY LIQUID IN TIME DOMAIN**

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Experimental evidence related to dielectrical response of different nematic and smectic A as well as viscous adhesives will be shown in brief. The response is quantitatively the same. There is possible to discriminate the bulk effects and electrode effects as well as AC and DC transport conductivity.

Time domain technique can be also used for investigation of crosslinked adhesives and solidified liquid crystalline materials.

Another group of problems that can be investigated by time domain technique is the influence of electrode shape and material.

The results can be compared with that of frequency domain technique. It is suggested that time domain technique is well suited at lowest frequencies where the influence of the electrode properties are of importance. This may be of interest to those investigating material ageing phenomena.

Also, reported experimental tests are of importance in respect to the use of time domain in dielectric tracking of reaction kinetics.

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## TIME DOMAIN TRACING ON CROSSLINKING REACTION IN EPON 828-EDA STECHIOMETRIC MIXTURE

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There are considerable amount of papers related to the investigation of crosslinking reaction kinetics by means of dielectric spectroscopy. Almost all of them were done in frequency domain at frequencies 10 kHz or less. In numerous papers the use of comb electrodes was reported.

The paper is linked to the use of time domain instead of frequency domain technique. The advantages of this technique are as following:

i/ it may deal with sample nonlinearities

ii/ bulk AC and DC conductivity as well as electrode phenomena can be separated in majority of cases.

The range of times explored is equivalent to indicated above frequency range.

It is shown that the dielectric response to the reaction is dependent not only on the frequency but also on the kind of the electrode used.

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## **INVESTIGATION OF ION MOBILITY BY MEANS OF TIME-OF-FLIGHT METHOD IN DGEBD-4D EPOXY-AMINE SYSTEM UNDER CURING**

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Dielectric techniques are widely used for *in situ* monitoring of resin curing. The basic assumption in these techniques is that the ionic conductivity determined from such experiments is directly related to the medium viscosity. It means it is assumed, that the ion concentration is constant and only ion mobility decreases in a course of the reaction. The Time-of-Flight (TOF) concept was recently proposed for direct determination of ion mobility in the model epoxy resin system diglycidylether of bisphenol A (DGEBA) with diamino4,4'dimethyl-3,3'dimethyldicyclo-hexylmethane (3DCM) [1,2]. It was found, that during curing, before the gelation, the ionic conductivity decreases and the time-of-flight of the ionic carriers increases so, that the ionic conductivity dependence versus ionic mobility is linear. However when the system was approaching the gelation the decrease of the ionic conductivity becomes faster than that of mobility, indicating that also some decrease of the ion concentration occurs. Unfortunately for the above system it was not possible to perform TOF experiments during the entire curing process because of the vitrification phenomena and low mobility of the ions in the medium ( $\sim 10^{-9}$  cm<sup>2</sup>/Vs at the beginning of the curing).

In the present work the TOF method is applied for the ion mobility measurements in another epoxy-amine system, diglycidylether of 1,4-butanediol (DGEBD) and 4,9-dioxa1,12-dodecane-diamine (4D) under isothermal curing. This system is

characterised by low  $T_g$  (-50°C), thus no vitrification phenomena occurs during the curing. This system shows relatively high conductivity and high ion mobility before as well as after the gelation, thus allowing to perform the TOF experiments during the entire hardening process. The evolution of ion mobility was determined from the series of subsequent TOF measurements using ionblocking gold electrodes (the TOF experiments require ion-blocking electrodes). The observed changes of the ion conductivity and mobility are related to results obtained from other independent methods (DMA, DSC, dielectrometry DC measurements). It is found that before the gelation the ion mobility decreases from  $5 \cdot 10^{-5} \text{ cm}^2/\text{Vs}$  to  $2 \cdot 10^{-7} \text{ cm}^2/\text{Vs}$ . These changes are linear with an advancement of the reaction. There is also linear dependence between ionic conductivity and ion mobility before the gel point what means that decrease of the conductivity during the reaction results from the increase of the medium viscosity. On the other hand, after the gelation, the relationship between ionic conductivity and ion mobility is no more linear indicating, that the decrease of ionic conductivity results also from a decrease of concentration of mobile ions.

We conclude, that the evolution of ionic conductivity can be used as a measure of the conversion before the gelation; however above the gelation the applicability of electrical methods for monitoring of resin curing is not obvious.

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## **DIELECTRIC SPECTROSCOPY OF BLENDS OF LIQUID CRYSTALLINE CELLULOSE DERIVATIVES**

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Several cellulose derivatives like (hydroxypropyl)cellulose (HPC) ((propionyloxy)propyl)cellulose (PPC) and ((hexanoyloxy)propyl)cellulose (HxPC) form lyotropic liquid crystalline solutions in the acrylic acid (AA). The lyotropic solutions can be oriented by mechanical shearing and then the orientation can be fixed by photo-induced polymerization of the AA. In such a way we have obtained highly anisotropic polymer networks showing strong birefringence and characteristic band texture [1,2]. Molecular relaxation phenomena in the polymer films were investigated by dielectric spectroscopy in a frequency range 20 Hz -1 MHz. PPC/pAA blends show the dielectric relaxation processes at the same temperature and with the same activation energy as pure PPC with superposition of the relaxation peak of pure pAA. Similar behaviour was found for the HxPC/pAA blends. The dielectric spectra of the HPC/pAA blends are strongly modified as compared with the spectra of HPC and pAA: the loss maxima are narrower and shifted towards higher temperatures (at constant frequency) or towards lower frequencies (at constant temperature). The differences in the behaviour of the HPC/pAA blends as compared with PPC/pAA or HxPC/pAA blends were found also in the Thermo-Optical Analysis (TOA), DSC and X-ray investigations. The isotropisation temperature of the HPC in the blends with pAA is strongly increased in comparison with pure HPC (above the thermal degradation temperature), while for the PPC/pAA or HxPC/pAA blends these temperatures are practically the same as for pure PPC or HxPC, respectively.

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These investigations indicate that in the HPC/pAA blends the intermolecular interactions between the components are much stronger than in the case of the PPC/pAA and HxPC/pAA blends. It is caused most probably by a formation of the hydrogen bonds between -OH groups from HPC and -COOH groups from pAA and afterwards by crosslinking of the polymer chains. It is concluded that intermolecular interactions in the investigated blends contribute to an increase in the thermal stability of the birefringence in the anisotropic blends.

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## **A STUDY OF METAL/TFREMO/METAL STRUCTURES BY MEANS OF THERMALLY STIMULATED CURRENTS**

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Thermally stimulated processes, namely: thermally stimulated currents (TSC), thermally stimulated electron emission (TSEE), thermally stimulated capacitance (TSCAP) or thermally stimulated luminescence (TSL) are powerful tools for examinations of trapping centers in semiconductors or insulators. In previous papers the dielectric data and dielectric response for thin films of rare earth metal oxides (TFREMO) sandwiched between metallic electrodes were examined by us in the frequency and time domain.

In this paper the TSC method is applied for electrical studies of vacuum deposited MIM structures which include some of TFREMO (sesquioxides of ytterbium, holmium, dysprosium and gadolinium) as representatives of a wider group the "rare earth family". TFREMO were prepared by electron gun deposition method. Their thickness was in the range 50nm-500nm. All electrical measurements were carried out for M/TFREMO/M thermoelectrets with aluminium electrodes in the temperature range from 297K to 500K. The influence of the polarising temperature, time and voltage as well as the heating rate on the TSC characteristics was tested.

The TSC thermograms obtained for these M/TFREMO/M structures at different measurement conditions were very similar. Only a single peak located at

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temperatures about 380K-410K was observed in the experimental thermograms. The nature of the observed peaks of thermally stimulated depolarisation (TSD) and thermally stimulated polarisation (TSP) for different rare earth oxides seems to be very similar. The TSC response in the temperature region 297K-500K is connected with trapping centers in TFREMO situated at energy depth of about 0.5-0.7eV, as determined with different estimation methods.

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## **TEMPERATURE INCREMENT INDUCED BY MAGNETIC FIELD IN SOME NEMATICS**

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Moll and Ornstein and later Miesowicz and Jezewski have observed that the temperature changes in nematic mesophases change as a result of magnetic fields. This phenomenon have been considered as a result of ordering produced by the interface and magnetic field.

Yun and Fredrickson carried out analogous experiments with a large sample volume for two compounds: PAA and DBA. They have found that the temperature increase due to the application of the magnetic field is permanent rather than transient and the sample returned to the surrounding temperature when the field was removed. They explained this continuos heat generation in a nematic sample by the magnetic field as a results of competition of magnetic forces and thermal motions exerted on the molecule.

In this work analogous experiments carried out for three nematics compounds: PAA, MBBA, EBBA are described. The magnetic field used was  $B=0.2\text{T}$ . For all these compounds the temperature rises were observed (the maximum value : 0.5K, 0.15K, 0.7K respectively ), but the shape of the temperature characteristics of temperature changes was different for all the cases.

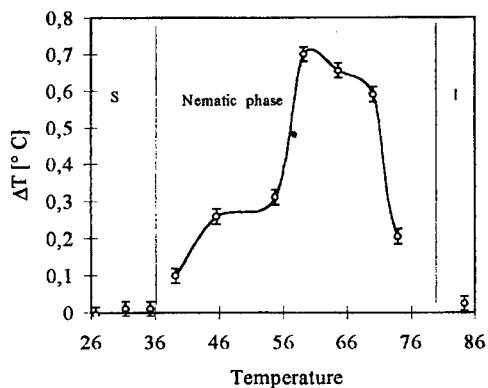


Fig.1. Temperature change  $\Delta T$  as a function of sample temperature. EBBA sample. Applied magnetic field  $B=0.2T$ .

We explain this phenomenon as a results of anisotropy of thermal conductivity, existing in nematics. When the magnetic field is applied to a nematic sample, the molecules are ordered along the field, so the anisotropy of the thermal conductivity takes place. In this situation the heat transport ( in geometry of our experiment ) from the heater to the thermocouple was better and the temperature measured was higher. The interpretation was tested by the measurements on MBBA at the room temperature. When both the nematic sample and the heater are at the same temperature ( this is possible for MBBA in the room temperature) no heat generation at the magnetic field has been detected.

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## **INVESTIGATION OF LIQUID DIOLS AND THEIR SOLUTIONS WITH DIELECTRIC SPECTROSCOPY**

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One of the important fields of the dielectric studies is the investigation of the intra- and intermolecular interactions in rather complicated liquids and their solutions. Unfortunately the dielectric theory of such substances is not quite evolved.

We believe that to establish a general dielectric theory of liquids and solutions it is necessary to measure dielectric properties of liquids with different types of intra- and intermolecular interactions. The wide-spread variety of these interactions is the hydrogen bonding.

There are some theoretical relations between the experimental results and the concept of the liquid structure for substances with one hydrogen bonded center (monoalcohols, phenols, monoamines, etc.). But making the conclusions on the basis of dielectric investigation about the structure of highly associated liquids with several hydrogen bonded centers is much more difficult problem.

In this work we discuss our experimental results for a number of diols (dialcohols), namely: 2-methyl-2,4-pentanediol, 2,5-hexanediol, 1,7-heptanediol, and 2,2-dimethyl-1,3-propanediol and also for the solutions of 1,3-propanediol in benzene. The measurements of dielectric relaxation were made in the wide range of temperatures and frequencies.

The obtained spectra may be described by Davidson-Cole equation, and in some cases as the sum of Debye type equations. The relaxation processes are connected with reorganization of hydrogen bonded network.

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## **DIELECTRIC RELAXATION AND MOLECULAR MOTION IN PC-BASED MICROEMULSIONS**

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**Ido Nir and Yuri Feldman**

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Time domain dielectric spectroscopy was applied to study molecular motion in w/o microemulsions composed from Soy-bean Phosphatidylcholine (SPC), water and oil (tricaprylin + n-hexanol in 1:1 molar ratio). The composition was chosen to give solutions of reversed micelles in oil medium. Such system where water and hydrocarbon phases are separated by surfactant molecules (SPG) is very interesting by simplicity of preparing microdroplets with different form and size.

Dielectric study was made in the frequency range 100 kHz - 5 GHz. Several relaxation processes were found. The low-frequency process with relaxation time about 1000 ns is attributed to rotation of micelles in oil medium. Fluctuational dipole moment of micelle is calculated equal to several dozens of Debye and is caused by fluctuation of individual SPC molecules in the micelle shell. Another two processes are characterized by relaxation times from one to several nanoseconds. They are attributed to (a) rotational motion of SPC molecules about their long axis and (b) rotation of the SPC polar heads in water core of micelles. Dipole moments of different parts of SPC molecules are estimated and compared with geometrical ones.

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## **ELASTIC PROPERTIES OF NEMATIC 4-[TRANS-4'-N-ALKYLCYCLOHEXYL] ISOTHIOCYANATOBENZENES**

**B. Żywucki, J. Jadżyn and G. Czechowski**

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Poznań Smoluchowskiego 17, Poland*

Elasticity is a fundamental test for application capability of nematic liquid crystals. In order to determine components of elasticity tensor we studied the Frederics transitions for thin layer of n-CHBT's, placed in external electric and magnetic fields in function of the temperature. From values of threshold fields  $E_c$  and  $B_c$  we determined the splay ( $K_1$ ), twist ( $K_2$ ) and bend ( $K_3$ ) elastic constants and anisotropy of magnetic susceptibility ( $\alpha$ ) of studied series. This macroscopic parameters show a characteristic alternation as a function of the number of aliphatic chain segments (odd-even effect).

Used the Landau-de Gennes model, we show the temperature dependence of the elastic constants as a combination of order parameter  $S$  (determined from  $\Delta\chi$  values) and parameters of expansion  $\alpha$  of free energy towards  $S$ . From the temperature dependence of  $S$  and  $K_i$  we determined parameters of expansion of free energy operator in nematic phase for n-CHBT series.

---

## ELASTIC CONSTANTS OF 6-TPEB

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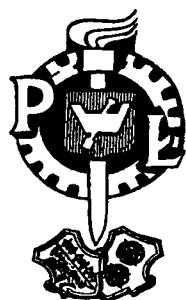
The Frederics transition of the 1-[3-6-alkyl-biphenyl]-2-[isothiocyanatophenyl]-ethane (6-TPEB) thin layer placed in external magnetic and electric field as a function of temperature have been studied. The temperature dependence of the splay and bend elastic constants have been determined. The anisotropy of magnetic susceptibility  $\Delta_\chi$  of 6-TPEB has been determined. From the value of birefrigerence and  $\Delta_\chi$ , the temperature dependence of order parameter has been found.

In the vicinity of nematic-smectic B phase transition the elastic constants have shown the pretransitional behaviour. On the basis of Jahling theory, the values of critical exponents obtained from temperature dependence of elastic constants in the vicinity of nematic-smectic B phase transition has been discussed.

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IV-th International Conference on  
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16 - 20 September  
Szczyrk, POLAND

**Technical University of Łódź,  
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**DIELECTRIC AND RELATED PHENOMENA**

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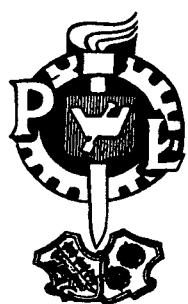
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IV-th International Conference on  
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16 - 20 September  
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The Conference is organised under the honorary Chairmanship of  
**prof.dr hab. Aleksander Łuczak**  
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## SCIENTIFIC PROGRAMME

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**Monday, 16<sup>th</sup> September**

### **Morning Session**

9 <sup>15</sup> - 9 <sup>30</sup>	<i>Opening of the Conference</i>
9 <sup>30</sup> - 10 <sup>30</sup>	<b>D.Lacey (UK)</b> - <i>Side chain liquid crystalline polymers</i>
10 <sup>30</sup> - 11 <sup>30</sup>	<b>Y.Séguí (France)</b> - <i>Surface flow on isolators</i>
11 <sup>30</sup> - 12 <sup>00</sup>	<i>Coffee break</i>
12 <sup>00</sup> - 12 <sup>30</sup>	<b>M.Stockhausen (Germany)</b> - <i>Dielectric spectroscopy of electrolyte solutions: a comparison of the lower frequency (ionic) relaxation contribution in various polar solvents</i>
12 <sup>30</sup> - 13 <sup>00</sup>	<b>J.J.Moura Ramos (Portugal)</b> - <i>Some comments on the physical significance of the compensation behaviour associated to the glass transition relaxation as observed by thermally stimulated currents</i>
13 <sup>00</sup> - 15 <sup>00</sup>	<i>Lunch</i>

### **Afternoon session**

15 <sup>00</sup> - 16 <sup>00</sup>	<b>T.Pajkossy (Hungary)</b> - <i>Admittance of surface with fractal surfaces</i>
16 <sup>00</sup> - 16 <sup>30</sup>	<b>J.Ulański (Poland)</b> - <i>Dielectric spectroscopy of blends of liquid crystalline cellulose derivatives</i>
16 <sup>30</sup> - 17 <sup>00</sup>	<b>B.Hilczer (Poland)</b> - <i>Role of the proton surroundings in fast proton transport</i>
17 <sup>00</sup> - 17 <sup>30</sup>	<b>B.S.Saburov (Tajikistan)</b> - <i>Dielectric relaxation in liquid crystal mixtures (solutions) with component molecules of different asymmetry of shape</i>
17 <sup>30</sup> - 18 <sup>00</sup>	<b>J.Małecki (Poland)</b> - <i>The relaxation of the nonlinear dielectric effect</i>
19 <sup>00</sup>	<i>Conference dinner</i>

## Tuesday, 17<sup>th</sup> September

### Morning Session

9<sup>00</sup> - 10<sup>00</sup> K.Weron (Poland) - *Arrow of time, probability and relaxation*  
10<sup>00</sup> - 11<sup>00</sup> S.Gubański (Sweden) - *Insulation life*  
11<sup>00</sup> - 11<sup>30</sup> *Coffee break*  
11<sup>30</sup> - 12<sup>00</sup> Z.Gburski (Poland) - *Application of the crystallisation process in glass-forming polymers*  
12<sup>00</sup> - 12<sup>30</sup> G.I.Ovtchinnikova (Russia) - *Ionic transport in order-disorder ferroelectrics via high frequency dielectric dispersion and nmr*  
12<sup>30</sup> - 13<sup>00</sup> J.Rybicki (Poland) - *Influence of doping on the conductivity of thin dielectric layers: a Monte Carlo study*  
13<sup>00</sup> - 15<sup>00</sup> *Lunch*

### Afternoon session

15<sup>00</sup> - 15<sup>30</sup> V.Novikov, O.Poznansky (Ukraine) - *Dispersion dependencies of dielectric properties. Renormgroup's approach.*  
15<sup>30</sup> - 16<sup>00</sup> Y.O.Roizin (Ukraine) - *Improvement of dielectric properties on silicon dioxide surfaces after processing in hexamethyldi-silizane*  
16<sup>00</sup> - 16<sup>30</sup> A.B.Szymański (Poland) - *Dielectric response of viscous and glassy liquids in time domain*  
16<sup>30</sup> - 17<sup>00</sup> G.W.Bąk (Poland) - *Dielectric relaxations in organic carrier-dominated systems*  
17<sup>00</sup> - 17<sup>30</sup> P.Adamski (Poland) - *Study of  $\alpha_{||}$  and  $\alpha_{\perp}$  of cholesteryl nonanoate molecule*

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17 <sup>30</sup> - 18 <sup>00</sup>	<b>M.Kryszewski</b> (Poland) - <i>The use of lateral impedance spectroscopy for evaluation of polymer coatings in microelectronics</i>
19 <sup>00</sup>	<b>Dinner</b>
20 <sup>00</sup> - 21 <sup>00</sup>	<b>Poster session</b>

### **Wednesday, 18<sup>th</sup> September**

8<sup>30</sup> - 13<sup>00</sup> **Excursions - Pszczyna**

13<sup>00</sup> - 15<sup>00</sup> **Lunch**

#### **Afternoon session**

15 <sup>00</sup> - 16 <sup>00</sup>	<b>P.A.Rolla</b> (Italy) - <i>Dielectric relaxation phenomena in polymers: recent findings by chemical vitrification experiments</i>
16 <sup>00</sup> - 16 <sup>30</sup>	<b>D.Fioretto</b> (Italy) - <i>Dynamics of polymers near the glass transition studied by Brillouin and dielectric spectroscopy</i>
16 <sup>30</sup> - 17 <sup>00</sup>	<b>T.Perova</b> (Russia) - <i>Electric field induced rotations and relaxation dynamics of FLC's studied by infrared, dielectric and optical methods</i>
17 <sup>00</sup> - 17 <sup>30</sup>	<b>S.J.Rzoska</b> (Poland) - <i>Relaxation of critical fluctuations in a binary solution. Nonlinear dielectric effect studies</i>
17 <sup>30</sup> - 18 <sup>00</sup>	<b>A.M.Ghoneim</b> (Egypt) - <i>Dynamic dielectric properties of carboxylic acid esters in benzene solution</i>

19<sup>00</sup> **Dinner**

21<sup>00</sup> **Barbecue**

## Thursday, 19<sup>th</sup> September

### Morning session

9 <sup>00</sup> - 10 <sup>00</sup>	<b>R.Richert</b> (Germany) - <i>Dielectric relaxation of the electric field in polyvinylacetate</i>
10 <sup>00</sup> - 11 <sup>00</sup>	<b>C.Laurent</b> (France) - <i>Optical pre-breakdown warnings in insulators</i>
11 <sup>00</sup> - 11 <sup>30</sup>	<i>Coffee break</i>
11 <sup>30</sup> - 12 <sup>30</sup>	<b>A.Ziabicki</b> (Poland) - <i>Effects of intermolecular interactions and external fields on orientationalphase transitions in systems of axially symetric, rigid particles</i>
13 <sup>00</sup> - 15 <sup>00</sup>	<i>Lunch</i>

### Afternoon session

15 <sup>00</sup> - 16 <sup>00</sup>	<b>G.W.Bąk, M.Kryszewski</b> (Poland) - <i>Low frequency dielectric properties</i>
16 <sup>00</sup> - 16 <sup>30</sup>	<b>Y.Feldman</b> (Israel) - <i>Time domain dielectric spectroscopy. Theory and applications.</i>
16 <sup>30</sup> - 17 <sup>00</sup>	<b>L.Murawski, R.J.Barczyński</b> (Poland) - <i>Comparison of KWW and power law analyses in iron phosphate glasses</i>
17 <sup>00</sup>	<i>Closing of the Conference</i>
19 <sup>00</sup>	<i>Dinner</i>

## Friday, 20<sup>th</sup> September

8 <sup>30</sup>	<i>Excursion - Wieliczka</i>
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## LIST OF POSTERS

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- **A.Błachut, M.Michalski** (Poland) - *More on comb electrodes*
- **A.Bródka** (Poland) - *Properties of liquid acetone in silica pores: molecular dynamics simulation*
- **I.Danielewicz-Ferchmin** (Poland) - *Density of water in high electric fields*
- **Z.Dendzik** (Poland) - *Scaling properties of glass-forming liquids*
- **A.Drozd-Rzoska** (Poland) - *Critical behaviour of electric permittivity in the isotropic phase of nematogens*
- **I.V.Ermolina** (Russia) - *Application of TDS for investigation of aqueous protein solution*
- **K.Friedrich** (Poland) - *Investigation of ion mobilin by means of time-of-flight method in DGEBD-4D epoxy-amine system under curing*
- **M.Górny** (Poland) - *A new application for studying nonlinear dielectric effect in liquids*
- **P.Habdas** (Poland) - *High pressure studies of electric permittivity near a critical consolute point in binary solutions*
- **J.Hoffman** (Poland) - *Simple model of the dielectric properties of cholesteric liquid crystals*
- **J.Jadżyn** (Poland) - *Elastic properties of nematic 4-(trans-4'-n-alkylcyclohexyl) isothiocyanobenzenes*
- **L.Jarecki** (Poland) - *Kinetic model of crystal nucleation in polar polymers in electric field*
- **A.Kocot** (Poland) - *The dynamics of the ferroelectric Lcs in helical and surface stabilised cells. Dielectric and electro-optical study and a computer model*
- **F.Kuliesius** (Lithuania) - *Regularities of electret charge relaxation in  $Zn_{0.7}C_{0.3}S$  layers*
- **M.Lucchesi** (Italy) - *Dielectric response dominated by charge transport in the poly(3-n-decylopyrrole) films*
- **M.Mucha** (Poland) - *Electro-optic and dielectric behaviour of PDLC films*
- **M.Olszowy** (Poland) - *Piezoelectricity and dielectric properties of PVDF/BaTiO<sub>3</sub> composites*
- **G.I.Ovtchinnikova** (Russia) - *Low frequency dielectric dispersion and universal law in ferroelectric polymers and ionomers*

- **M.Paluch** (Poland) - *Dielectric relaxation of glass-forming liquids under high pressure*
- **K.Pasterny** (Poland) - *MD simulation study of a liquid crystal model: Gay-Berne particles with dipole moments*
- **A.Rybicka** (Poland) - *Polaron transport properties derived from MD-simulated structure in V<sub>2</sub>O<sub>5</sub>-P<sub>2</sub>O<sub>5</sub> glass*
- **J.Rybicki** (Poland) - *Transient currents in thin layers for simultaneous multiple-trapping and hopping transport mechanisms*
- **J.Rybicki** (Poland) - *The influence of the modified hopping centre density in the near-contact regions on the TOF currents*
- **S.Różański** (Poland) - *Determination of the order parameter in nematic liquid crystal confined to anopore membranes*
- **E.Rysiakiewicz-Pasek** (Poland) - *Influence of water on thermally stimulated current in porous glasses*
- **B.S.Saburov** (Tajikistan) - *Electrophysical properties of liquid crystal in dynamics of X-ray irradiation in the range of temperatures of phase transitions*
- **B.Schiener** (Germany) - *Dynamic heterogeneity in supercooled liquids revealed by non-resonant dielectric spectral hole burning*
- **O.I.Shpotyuk** (Poland) - *Relaxation structural transformations in a-As<sub>2</sub>S<sub>3</sub> associated with radiation-induced centers*
- **E.Szwajczak** (Poland) - *On some processes in liquidcrystalline electrolytes*
- **A.Ł.Szymański** (Poland) - *Time domain tracing on crosslinking reaction in epon 828-EDA stochiometric mixture*
- **T.Wiktorczyk** (Poland) - *A study of metal/tfremo/metal structures by means of thermally stimulated currents*
- **M.Wojciechowski** (Poland) - *Temperature increment induced by magnetic field in some nematics*
- **V.I.Zhuravlev** (Russia) - *Hydrophobic effect and dielectrical and optical properties of nonelectrolyte aqueous solutions*
- **V.I.Zhuravlev** (Russia) - *Investigation of liquid diols and their solutions with dielectric spectroscopy*
- **Y.Zuev, O.Zueva** (Russia) - *Dielectric relaxation and molecular motion in PC-based microemulsions*

## **GENERAL INFORMATION**

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All session of the IV-th International Conference will take place at the Hotel „WŁÓKNIARZ” in Szczyrk near Bielsko-Biała.

### **The Conference Office**

will be open from Sunday, 15<sup>th</sup> September at 12<sup>00</sup>, throughout the Conference.

### **Registration**

The registration desk, at the Hotel „WŁÓKNIARZ” in Szczyrk will be open from Sunday, 15<sup>th</sup> September 1996 at 12<sup>00</sup> and during the Conference hours.

Registration includes admission to all scientific session, meeting materials, meals and accommodation.

### **Meals**

All meals will be served at the Hotel „WŁÓKNIARZ”:

<i>Breakfast</i>	at	8 <sup>00</sup>
<i>Lunch</i>	at	13 <sup>00</sup>
<i>Supper</i>	at	19 <sup>00</sup>

Coffee, tea and soft drinks will be served during the breaks of the scientific programme.

### **Social events**

All participants are invited to the Conference Dinner on Monday, 16<sup>th</sup> September at 19<sup>00</sup>.

### **Excursion**

Wednesday, 18<sup>th</sup> September - Pszczyna  
Friday, 20<sup>th</sup> September - Wieliczka

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# **DIELECTRIC AND RELATED PHENOMENA**

## **DRP'96**

### **Abstracts**

### **SUPLEMENT**

Szczyrk, Poland  
16 - 20 September 1996

TECHNICAL UNIVERSITY OF ŁÓDŹ  
BRANCH IN BIELSKO-BIAŁA

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**DIELECTRIC AND RELATED PHENOMENA**

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## CRITICAL BEHAVIOUR OF DIELECTRIC PERMITTIVITY IN THE ISOTROPIC PHASE OF NEMATOGENS

A. Drozd-Rzoska, S.J. Rzoska, J. Ziolo

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It has been shown that the temperature behaviour of dielectric permittivity ( $\epsilon$ ) in the isotropic phase of nematogens can be described in the same way as in critical binary solutions. Hence using a relation (1) it was possible to portray the results of  $\epsilon(T)$  measurements in the isotropic phase of 5-heptyl-2-(4'cyanobiphenyl)-pyrimidine and 4,4-n-octycyanobiphenyl:

$$\epsilon(T) = \epsilon^* + a^* \times (T - T^*) + A^* \times (T - T^*)^\phi \quad (1)$$

where:  $\phi = 1 - \alpha \approx 0.5$ ,  $T^* = T_{I-N} - \Delta T$  and  $T > T_{I-N}$ ;  $\alpha$  is the critical exponent the specific heat,  $T_{I-N}$  denotes the isotropic - nematic (I - N) phase temperature.

The influence of the position of the permanent dipole moment on results was tested by additional measurements in n-(p-methoxybenzylidene)-p'butylaniline.

It also has been shown that the fluidlike analogy can be applied to nonlinear dielectric effect (NDE) which describes changes of electric permittivity induced by a strong electric field. Measurements were conducted for the lowest frequency used in NDE studies,  $f = 67$  kHz, so the condition:

$$f^l \gg \tau \quad (2)$$

where:  $\tau$  - denotes relaxation frequency of pretransitional processes was always fulfilled.

Values of discontinuities of the isotropic - nematic phase transition ( $T_{I-N} - T^*$ ) from both the analysis of  $\epsilon(T)$  measurements and NDE studies are in good agreement.

It seems that the presented analysis may offer an adequate description common description of the pretransitional  $\epsilon$  and NDE effect. Recently, the fluidlike analogy has been successfully theoretically investigated by Mukherjee et al. (*Phys. Rev. B* **52**, 9964 (1995)), which pointed out the possibility of critical behaviour with the dimensionality  $d=3$  for the I-N phase transition.

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## **A NEW APPLICATION OF NONLINEAR DIELECTRIC METHOD FOR STUDYING RELAXATION PROCESSES IN LIQUIDS**

**M.Górny, J. Ziolo and S.J. Rzoska**

*Institute of Physics, Silesian University, ul. Uniwersytecka 4,  
40-007 Katowice, Poland*

The measurement set - up for studying in liquids changes of electric permittivity induced by a strong electric field (nonlinear dielectric effect, NDE) is presented. The construction is based on the idea of frequency modulation of LC generator, proposed by Małecki [J. Chem. Soc. Faraday Trans. II, 72, 104 (1976)]. The strong electric field is applied in the form of rectangular pulses (typically 1 4 ms). The set up enables measurements in a broad range of frequencies (80 kHz - 12 Mhz) and contains a new calibrating system, minimising the influence of a systematic error in the measured NDE values. The digitising system with custom software makes measurements under heavy noisy conditions possible.

It is shown the application of the presented apparatus for time-resolved NDE studies of relaxation processes in liquids.

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## **KINETIC MODEL OF CRYSTAL NUCLEATION IN POLAR POLYMERS IN ELECTRIC FIELD**

**L. Jarecki**

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Application of an electric field to a polar polymer results in crystallisation of oriented polar phase. Model calculations confirm empirical conclusions that electric field induces nucleation and growth of oriented polar crystals, e.g. in PVDF. The mechanism of polar phase crystallisation is not of the type of classical oriented crystallisation from pre-oriented amorphous matrix, but it is related to orientation-dependent electrostatic potential energy of the resulting polar crystals in the field.

Model calculations indicate that primary nucleation rate, and growth rate of a polar crystal, both depend on orientation of the crystal in electric field. The effects of electric field are predicted to be very sensitive to the degree of undercooling. For instance, to keep field effects at a constant level, field intensity should increase nearly proportional to the third power of undercooling. At a high degree of undercooling, field effects to be detected may require electric field intensities far beyond the discharge level.

The model predicts also a weak decrease of nucleation and growth rates for the case of a polar crystals in the field.

Example computations of electric field effects on nucleation and growth rates of polar crystals are performed and illustrated for PVDF.

## **REGULARITIES OF ELECTRET CHARGE RELAXATION IN $Zn_{0.7}Cd_{0.3}S$ LAYERS**

**F. Kuliesius, S. Tamaiunas, A. Pindulis**

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2054, Lithuania*

Transient processes in  $Zn_{0.7}Cd_{0.3}S$  layers (which, in composition with ferroelectric liquid crystal, are used as the light modulators) are strongly affected by the electret charge.

The peculiarities of electret charge information as well as the electret voltage dependences on poling electric field, poling time and temperature have been analysed in  $Zn_{0.7}Cd_{0.3}S$  layers during the present work. The mechanism of space charge accumulation have been determined. The applications of the obtained results for the adjustment of the conditions of ferroelectric liquid crystal based light modulators are presented, also.

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## SIDE CHAIN LIQUID CRYSTALLINE POLYMERS

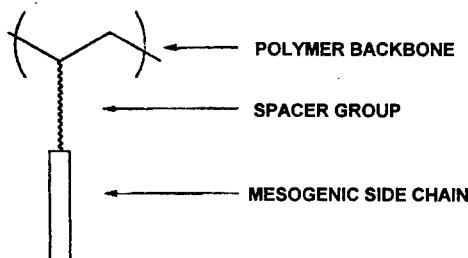
D. Lacey

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United Kingdom*

Side chain liquid crystal (SCLC) polymers, as its name suggests, involves the attachment of mesogenic moieties as side chains to the polymer backbone. Most polymer backbones will give a random arrangement of the chain segments and so if a polymer backbone is stretched out due to the mesophase ordering, then it will lose a lot of its entropy. Conversely, the mesophase moiety has a strong tendency towards the anisotropic arrangement of the molecules and any distortion of the mesophase field by the random nature of the polymer backbone will result in a large mesophase elastic energy. Thus the polymer entropy is antagonistic towards mesophase ordering.

The incompatibility of polymer entropy and mesophase ordering is somewhat ameliorated by the introduction of a flexible spacer group, which links the mesogenic side chain to the polymer backbone. The decoupling, or partial decoupling, of the mesogenic side chain by the flexible spacer group allows the polymer backbone motions to occur without disturbance of the anisotropic arrangement of the mesogenic side chains. The introduction of the flexible side group (our flexible friend) allows the SCLC polymer to exhibit a wide range of liquid crystalline phases.

This lecture will outline the main structural features of SCLC polymers, i.e. the polymer backbone, the spacer group and mesogenic side chain, and show how these can be manipulated to give a variety of different polymer architectures for use in a number of applications.



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## **ELECTRO-OPTIC AND DIELECTRIC BEHAVIOUR OF PDLC FILMS**

**M. Mucha, E. Nastal**

*Technical University of Łódź, Łódź*

**J. Jadzyn, G. Czechowski**

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Polymer Dispersed Liquid Crystals have attracted great attention not only for their physical properties but also for a wide range of possible applications. They consist of nematic liquid crystals randomly dispersed as droplets in polymeric film. PDLC films for the electrooptic and dielectric studies were prepared by blending appropriate amounts of a nematic liquid crystal mixture (LC, W-52) and the polymer precursor unsaturated oligoester resin containing styrene or diacrylane and a UV curing agent. The PDLC films of 10 to 40% of LC were obtained by LC separation during UV polymerization of the matrix. Thermo and electrooptical effects of the films were measured using polarised light a helium-neon laser.

Dielectric measurements were analysed over a log. frequency range 5-7 in temperatures 25-40 °C for various concentrations of LC. The broad experimental loss factor curves were fitted by four Debye's distribution functions. The values of log relaxation time are found to be independent on LC concentration in the PDLC systems. The early results suggest the well purity of LC in the droplets and very weak surface effect of the polymeric matrix on the LC dynamics in PDLC.

# **EFFECTS OF INTERMOLECULAR INTERACTIONS AND EXTERNAL FIELDS ON ORIENTATIONAL PHASE TRANSITIONS IN SYSTEMS OF AXIALLY SYMMETRIC, RIGID PARTICLES**

**A. Ziabicki, L. Jarecki**

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Potential flow, electric, or magnetic fields applied to a system of rigid interacting particles produce molecular orientation, affect physical properties of the system and influence orientational phase transitions.

Thermodynamics of orientational phase transitions in systems with dipole and quadrupole interactions, subjected to dipole and/or quadrupole external fields are analyzed. Different behaviour is predicted for energetic vs. entropic nature (e.g. Onsager excluded volume interactions).